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Evaluation of Relationship between Lead-Dust Loading, Lead-Dust Concentration, and Total Dust loading Metrics across multiple Data sets

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Abstract

Lead-dust monitoring studies report values as either lead-dust loadings µ*g/ft*^{2} or as lead-dust concentrations µ*g/g*. It is rare for studies to report both metrics. When only lead-dust loading values are present, professionals require an approach to estimate lead-dust concentration values. A literature search identified five studies that contained raw data for both lead-dust loading and lead-dust concentration. An additional thirty-two studies had summary-statistics available for both lead-dust loading and lead-dust concentration. Studies with raw-data were used to develop an empirically-based loading to concentration statistical relationship. Raw data sets were critically evaluated to determine whether elimination or substitution of data points was warranted. Studies with summary statistics were used to evaluate this relationship using independent data. Despite the differences in study-design, sampling method, and analytical procedures, the overall empirical relationship across studies with raw data remained consistent. The r^2 value between lead-dust loading and lead-dust concentration improved from 0.30 to 0.55 for raw data sets to 0.55 to 0.58 for pooled data sets. The standard error of the regression, slope, and intercept were also improved. Reported central-tendency summary statistics from independent data sets showed that measured central tendency dust lead concentrations from these studies were similar to predicted dust lead concentrations using the regression derived from raw data sets. Across 142 extracted paired lead-dust loading and lead-dust concentration pairs from independent data, >90% of estimated lead-dust concentration values were within an 80% prediction interval. The loading to concentration relationship developed from this data is log-log linear. The most likely reason is due to variable total dust loadings (g/ft^2), which result in one lead-dust loading value being linked to a range of lead-dust concentration values. When only lead-dust loading values are present, this empirical relationship provides a reasonable approach to estimate lead-dust concentrations.

Introduction

Lead-dust exposure remains a persistent and important contributor to elevated blood lead levels. Dust is a complex mixture of particles comprised of mineral and organic matter. Anthropogenic and naturally occurring chemical substances adhere to these particles through routine processes such as cooking, vacuuming, use of consumer products, and track-in of outdoor soil (Health Canada 2018). Lead is a ubiquitous and well-studied contaminant in dust (EPA 2019). Lead is consistently detected in dust in national surveillance studies, regional studies, intervention studies, and in historically contaminated sites (HUD 2011; Hunt et al., 2012; Wilson et al., 2006; Zota et al., 2016). Abrasion and flaking of lead-based paint, track in from contaminated soil, and use of consumer products have been reported as contributing sources to lead-dust (Gwiazda et al., 2000; Glorennec et al., 2010; Norman et al., 1997).

Few studies report all metrics (lead-dust loading, lead-dust concentration, and total-dust loading) when reporting lead-dust occurrence. All three metrics are positively correlated with each other. In this way, one lead-dust loading value can be associated with a range of lead-dust concentrations due to the range of total dust loadings present within and across different homes. Across the datasets considered here, the relationship between lead-dust loading and lead-dust concentration consistently appears as log-log linear. Incremental increases in lead-dust concentration occur faster at lower lead-dust loading levels and slow-down as lead-dust loading levels increase.

The two primary sampling methods for indoor dust are surface wipe and vacuum sampling. While both wipe and vacuum sampling can be used to measure lead-dust loading, wipes are more typically used. Vacuum samples can be used to measure lead-dust concentration, provided that enough dust was collected to allow for sieving and analysis. Total dust loading, also referred to as dust loading, is defined as the total mass of dust present across a defined surface area and can be measured by either sampling method.

If continuous measured paired data for any two of these metrics are available, the third metric can be estimated. For example, Hunt et al. (2012) estimated lead-dust concentration from measured lead-dust loading and total dust loading values. Rasumussen et al (2013) estimated lead-dust loading from lead-dust concentration and total dust loading values. Some studies have attempted to estimate lead-dust concentration by pairing continuous lead-dust loading data with a single point estimate for total dust-loading or by using a static ratio between lead-dust loading and lead-dust concentration (LeBot et al., 2010; Glorennec et al., 2012; Gulson et al., 2018). However, this analysis will show that use of static ratios or single point estimates are not supported by the observed relationships, which are driven by the high variability of total dust-loading within homes, between homes, and over time.

Previous studies have explored the relationship between continuous paired lead-dust loading and lead-dust concentration data (Adgate et al., 1995; EPA 1998; EPA 1993; Hunt et al., 2012; Lanphear et al., 1995; Clayton et al., 1999; Rasmussen et al., 2013). These studies have been used on their own and pooled together to develop an empirically based statistical regression where a single lead-dust loading value can be used to estimate a range of lead-dust concentrations (EPA 2008; EPA 2010; EPA 2014; EPA 2019). The U.S. EPA used pooled data from three studies (EPA 1998; Rasmussen et al., 2013; Adgate et al., 1995) to derive an empirically-based lead-dust loading to lead-dust concentration regression (EPA 2019). This regression is used as a baseline when comparing results presented in this study.

The purpose of this study is to determine if the addition of new data sets (Lanphear et al., 1995; Clayton et al., 1999) as well as substitution and elimination of individual data points can improve the overall relationship between lead dust loading and lead dust concentration.

Methods

This study explores the relationship between lead-dust loading and lead-dust concentration through the following three questions:

- Does addition of new raw datasets change the underlying statistical empirical relationship between lead-dust loading and lead-dust concentration?
- Does substitution or elimination of individual data-points within raw data sets change the lead-dust loading to lead-dust concentration relationship?
- Does comparison with independent data obtained through a literature search support statistical empirical relationship observed between lead-dust loading and lead-dust concentration from raw data-sets?

Consideration of Raw Datasets-Statistical Data Analysis

Different lead-dust metrics are reported based on the objectives of a given study. When comparing reported levels to regulatory values such as dust-lead hazard standard and clearance levels, lead-dust loading values are required. When using blood lead models or deriving intake values (µg/day), lead-dust concentration values are required. The overall relationship between lead-dust metrics is described by the following set of equations:

$$Eq.1{:}\ Lead\ Dust\ Loading\,\frac{ug}{ft2} = Lead\ Dust\ Concentration\,\frac{ug}{g} \times Total\ Dust\ Loading\,\frac{g}{ft2}$$

$$Eq.2{:}\ Lead\ Dust\ Concentration\,\frac{ug}{g} = \frac{Lead\ Dust\ Loading\ ug/ft2}{Total\ Dust\ Loading\ g/ft2}$$

$$Eq.3{:}\ Total\ Dust\ Loading\,\frac{g}{ft2} = \frac{Lead\ Dust\ Loading\ \frac{ug}{ft2}}{Lead\ Dust\ Concentration\ \frac{ug}{g}}$$

When only lead-dust loading values are reported, professionals require an approach to convert these values into a lead-dust concentration. Equations 4, 5, and 6 can be used to estimate lead-dust concentration values when only lead-loading loading values are available (EPA 2019):

$$\text{Eq. 4: lead dust concentration}\,\frac{ug}{g} = (EXP(Intercept + Slope \times (\ln(Pb\ dust\ loading) - Avg(\ln\,(Pbload)))$$

$$Eq\ 5{:}\ \ln(\text{concentration})\,\frac{ug}{g} = (Intercept + Slope \times \ln(Pb\ dust\ loading) - avg(\ln(Pbload)) + Error\ Variance$$

The inputs for the loading to concentration regression are obtained by regressing raw paired data sets containing lead-dust loading and lead-dust concentration values using the add-on Regression function in Excel®. Five data sets with raw paired lead-dust loading and lead-dust concentration values were identified (Adgate et al., 1995; EPA 1998; EPA 1993; Lanphear et al., 1995; Clayton et al., 1999; Rasmussen et al., 2013). Each sample from each dataset was equally weighted for these regressions.

Loading to concentration regressions from raw data sets were derived through the following statistical procedure. From the raw data, each lead-dust loading and lead-dust concentration was transformed by taking the natural log. The average of the natural log of the lead-dust loadings across all data points was subtracted from each lead-dust loading. This re-centers the lead-dust loading data which was regressed with the natural log of the lead-dust concentration using the add-on Regression function in Excel® to calculate a predictive distribution as detailed in Qian (2010). The regression output was used to calculate a prediction interval/error variance by combing a T value with the standard error of the prediction. The standard error of the prediction uses the standard error of the regression, standard error of the slope, standard error of the intercept as shown in Equation 6:

$$Eq\ 6\ error\ variance = TINV(1 - confint, residdf) \times sqrt(stderr^2 + stderrint^2 + stderrslope^2 X(\ln(Pbload) - meanlnPbload)^2))$$

Where:
T value: two tailed t-test (TINV), using upper confidence interval and residual degrees of freedom

Standard Error of Prediction:
Stderr: Standard Error of Regression
Stderrint: Standard Error of Intercept
Stderrslope: Standard Error of Slope
Pbload: Lead-dust loading, variable, user-defined
MeanlnPbload- Mean natural log of lead-dust loading

The error variance term predicts the variability around the regression line, which shows that one ln(PbLoad) can be associated with a range of ln(PbConc). The error variance term shows that there is a given probability that a lead-dust concentration will lie within the prediction interval for a given lead-dust loading value (EPA 2019).

The values used in the baseline loading to concentration regression are shown in Table 1 and are provided for context. The regression analysis was completed for several iterations of data sets as described in Table 2. Supplementary File 1: Lead-Dust Loading to Concentration Calculator shows how inputs such as those shown in Table 1 can be obtained for any paired data set and then used to estimate lead-dust concentration values.

Table 1: Inputs required to estimate central tendency dust-lead concentration plus an upper and lower predicted lead-dust concentration from a given lead-dust loading value for the baseline loading to concentration regression

Input	Value
Pb (lead) dust loading and its natural log	User defined
Upper bound of prediction interval	User defined
Slope	0.421
Y intercept	5.148
Average (ln) Pb-dust loading value	0.881
Standard Error of Slope	0.009
Standard Error of Intercept	0.018
Standard Error of Regression	0.737
Residual Degrees of Freedom	1641

Substitution or Elimination of Individual Data points

The raw data for each study was critically evaluated to determine whether substitution or elimination of data points were warranted. Clean data sets are defined as those where substitution or elimination of data points occurred. Pooled data sets are defined as the combination of three or more data sets. Table 2 provides an overview of data sets used in the regression analyses.

Table 2: Description of data sets used in regression analyses

Data set(s)	Sample Size
Baseline-pooled (HUD 1990; Rasmussen et al 2013; Adgate 1995)	1643
HUD 1990 (raw)	312
HUD 1990 (clean)	301
Rasmussen et al., 2013 (raw)	1022
Rasmussen et al., 2013 (clean)	1010
Adgate et al., 1995 (raw)	312 of 444
Adgate et al., 1995 (clean)	394 of 444
Clayton et al., 1999 (raw)	244
Clayton et al., 1999 (clean)	215
Lanphear et al., 1999 (raw)	202
Lanphear et al., 1999 (clean)	190
Pooled (HUD 1990; Rasmussen et al 2013; Adgate 1995; Lanphear et al 1995; Clayton et al 1999)-raw	2174
Pooled (HUD 1990; Rasmussen et al 2013; Adgate 1995)-clean	1705
Pooled (HUD 1990; Rasmussen et al 2013; Adgate 1995; Lanphear et al 1995)-clean	1895
Pooled (HUD 1990; Rasmussen et al 2013; Adgate 1995; Clayton et al 1999)-clean	1920
Pooled (HUD 1990; Rasmussen et al 2013; Adgate 1995; Lanphear et al 1995; Clayton et al 1999)-clean	2110

Analysis of Individual Data sets

The HUD Survey data measured floor lead-dust loadings using a vacuum sampling method and used equations based on statistical relationships of other empirical data sets to estimate wipe-based lead-dust loading values (EPA 1993; EPA 1998). This is the oldest of the studies included in this regression. The blue nozzle method used to collect house dust is subject to lower vacuum collection efficiencies (EPA 2010). Total dust was collected through tapping dust from the blue-nozzle and lead-dust concentration was measured. The relationship between this blue-nozzle method and two other empirical data sets that used this approach as well as side-by-side wipe sampling provided the basis for estimation of wipe-based lead-dust loading values. A subsequent analysis of the data identified eight outliers through three different statistical models (EPA 1993). These outliers were noticeably extreme and were identified as having an inordinate effect on the data (EPA 1993). This study used the weighted average of three rooms to derive an average loading and average concentration value per house. In three instances, one or more of these rooms was not available so an imputed value was used. The clean version of the HUD data set removes these eleven data points.

A study by Adgate et al. (1995) examined the relationship between lead-dust loading, lead loading, and lead concentration in house dust in 216 homes in Jersey City, New Jersey. Wipe samples were collected for lead-dust loadings and were compared to lead-dust concentrations collected by a vacuum cleaner from wall to wall carpet and area rugs greater than 48 square feet. The total sample size was 444 data points for hard surface floors (Adgate et al, 1995). The data points from the original scatterplot in Figure 7A of Adgate et al., were digitized using the GetData®; because some of the data points were located on top of each other, only 394 of the 444 data points were captured during the digitization. Note, the baseline loading to concentration regression digitized 312 data points (EPA 2019). For this analysis, digitization was repeated three times and the digitization attempt with 394 data points achieved the closest match to reported summary statistics. For the all raw data regression, the digitization attempt with 394 data points was used.

The NHEXAS data set was collected in the mid-west region of the United States in the 1990s. The study was sponsored by the U.S. EPA and used the same sampling and analytical approach as the Adgate data (Adgate et al., 1995) (Clayton et al., 1999). The NHEXAS data set contained twenty data points that were below the level of detection for both lead-dust loading and lead-dust concentration. These values were removed from the analysis because of the uncertainty associated with estimating values below detection limits. The NHEXAS dataset also contained nine data points that were identified as outliers due to a very high-dust loading value as described in the section below.

The Rochester data set is described in several publications and was supported by the National Center for Healthy Housing (Lanphear et al., 1995) (Lanphear et al., 1997). The Rochester dataset contains data for 204 residences, and up to six rooms were sampled in each residence. One home was removed because no loading samples were present for any room, and another home was removed because only one concentration sample was present across the rooms. For the raw regression, geometric mean values for 202 data points were used instead of arithmetic mean values as these values are less subject to extreme values and produced an improved correlation (AM r^2=0.18, GM r^2=0.30). Of the remaining 202 samples, four additional samples were removed because they contained lead-dust loading or lead-dust concentration data for <3 rooms. For 89 homes, the number of rooms sampled for lead-dust loading and lead-dust concentration differed. To normalize, a surface-area weighted geometric mean was calculated. Weighting measurements by surface-area sampled when multiple samples are available is a common technique (HUD 2011). In this instance the surface area covered by each room was not available so 10% was assigned to the entryway and 90% was assigned to the rest-of-home samples. Using this metric, eight additional samples had a total dust-loading value >1.9 g/ft2 and were removed from the analysis, as informed by the overall outlier analysis presented below.

The Canadian House Dust Study (CHDS); (Rasmussen et al. 2013) collected lead-dust samples from 1,025 homes across 13 cities and was designed to estimate nationally representative urban house dust metal concentrations and metal loadings. Trained technicians followed a vacuum sampling protocol to collect a composite ("whole house") sample of active dust from all dry living areas of each house. Participants were instructed not to clean for at least one week prior to sample collection. The vacuum samples were sieved into fine (<80 µm) and coarse (80–300 µm) particle size fractions which were combined to calculate lead-dust loading values based on: measurements of the floor area sampled, dust mass, and lead-dust concentration (Rasmussen et al., 2013). As area measurements were not available for three houses, the overall sample size for the lead-dust and total dust loading calculation was 1,022.

Room by room wipe samples were also collected for 742 homes in the CHDS. The results for these homes are reported, as a completed dataset, for the first time here. A subset of these wipe samples was previously reported for three Canadian cities (McDonald et al 2010). The present study compares the CHDS wipe samples with CHDS whole-house vacuum samples for the lead-dust loading metric. The CHDS represents a unique data set where both room by room wipe sampling and whole-house vacuum sampling were used to report lead-dust loading for a large data set. Homes that met any of the following criteria were not included in this statistical comparison:

- only one-wipe collected per house (n=4),
- all wipes were below the detection limit (n=28), or
- only one sample was above the detection limit, but this sample was below the quantification limit (n=10).

This resulted in 700 of 747 homes with wipe samples that could be compared to the lead-dust loading values derived from whole-house vacuum sampling. Wipe samples were collected on hard surface flooring prior to whole-house vacuuming. Vacuum sampling collected dust from both hard surface floors and carpeted surfaces. Note, an additional 40 homes were flagged (as high uncertainty) due to low total-dust loading values as discussed below.

The comparison between wipe and vacuum data identified a small subset of samples where total dust loading values were exceptionally low. In these instances, a very small total dust mass (approximately one to two grams) was collected from the entire home. When this low total dust loading value was combined with a measured lead-dust concentration to estimate lead-dust loading, it resulted in considerable uncertainty. These estimated lead-dust loading values were below the level of detection (0.038 $\mu g/ft^2$) or level of quantification (0.128 $\mu g/ft^2$) that were applied to lead-dust loading using a wipe-based sampling method. Fifty-seven estimated lead-dust loading values were below this LOD or LOQ. Twelve of these had no matching wipe data and were removed from the clean CHDS data set. Five had wipe samples where all values were below the detection limit, and the vacuum lead-loading values were retained. The remaining forty samples had wipe-based loading values that were substituted into the clean CHDS data set as shown in Table 3.

Table 3: Comparison of Substituted Wipe Lead-dust Loading values and Vacuum Lead-dust Loading values for forty samples

Percent of samples	< LOD	>LOD and <LOQ	>LOQ and <1	Just above 1
Wipe sampling method	0%	25%	65%	10%
Vacuum Sampling method	5%	95%	0%	0%

Analysis across all Data sets

Raw data from all five data sets were used to conduct an outlier analysis using the total dust loading metric. The histogram of ln(total dust loading) is close to normal (not shown). Values greater than 2 times the interquartile range were identified as potential outliers. This translates to a value of 3.1 g/ft^2. Z scores were also calculated. A z-score of 3 translates to total dust loading value of 1.9 g/ft^2 and a z score of -3 translates to a value of 1E-4 g/ft2. The outlier analysis identified fourteen datapoints with high total dust-loading values >1.9 g/ft2. Two of the fourteen datapoints were previously identified as outliers from the HUD data set, nine of the fourteen datapoints were from the NHEXAS dataset, and three were from the Rochester Dataset. The analysis also identified one very low total dust-loading value from the Canadian House Dust Survey (CHDS). The data substitution procedure as described in Table 3 brings the total dust loading for this datapoint into the distribution.

The basis for removing or substituting values with very high or very low total dust loading values has a physical basis reflecting limitations in what can be measured. When using wipe-based method to measure total dust loading, Johnson et al., reported a LOQ of 0.005 g/ft^2 (Johnson et al., 2005). This is similar to the minimum value, 0.004 g/ft^2, reported in the CLEARS study (Adgate et al., 1995). Wipes are not as efficient at picking up dust, when compared to vacuums. They are used over smaller surface area and are not as durable on carpeted surfaces. Vacuum samplers generally collect more dust over a larger surface area, and studies have used this method to report total-dust loading values lower than 0.004 g/ft^2 and much higher values (Rasmussen et al., 2013) (Adgate et al., 1995) Vacuum-based sampling methods are more efficient at collecting dust. The sample flow volume and collection time influence how much dust is collected. Rasmussen et al., collected readily accessible surface or fresh dust using a high-efficiency vacuum cleaner using a light touch on carpets to avoid collecting deep dust (Rasmussen et al., 2013). For homes where there are reservoirs of dust imbedded within carpet, vacuuming this deep dust along with surface dust results in collection of more total dust which can unduly influence or dilute lead-dust concentration values in rare instances where total dust-loading values are very high (Roberts et al., 1999) (Yiin et al., 2002).

Identification of Previous Lead-dust Monitoring Studies

A literature search was conducted to identify studies where both lead-dust loading and lead-dust concentration values were reported. The literature search contextualizes reported lead-dust loading values by extracting central tendency summary statistics and grouping them by study type. It is notable that most studies report either dust lead loading or dust lead concentration, but not both. The literature search identified 2,310 data sources through the end of calendar year 2018.

Supplemental Information File 2 provides additional information on the literature search strategy and screening criteria (title and abstract and full-text) used to identify and screen lead-dust monitoring studies. In summary, studies did not pass screening if they did not meet any combination of the inclusion criteria. Common reasons that studies did not meet inclusion criteria were: they only measured external street or road dust, they did not contain quantitative monitoring data, or their sampling and analytical methods were not typical.

Supplemental Information File 2 also provides a brief overview of sampling and analytical methods typically used in lead-dust monitoring studies. Studies were excluded based on their sampling method if they measured dust accumulation or dust-fall through cups, trays, or dishes rather than measuring lead-dust loading and/or lead-dust concentration through wipes or vacuums. Studies were excluded based on their analytical methods if they had a very high level of detection (i.e., >10 µg/ft2), or measured lead-dust content through XRF rather than through traditional wet-chemistry analytical techniques. The studies used to develop and evaluate the loading to concentration relationship used different sampling methods (wipe versus vacuum) and contain spatial and temporal heterogeneity. The studies also used different analytical methods which resulted in different levels of sensitivity and variable detection and quantification limits.

Results

Summary of Data Sets Used to Develop Loading to Concentration Regression

Five data sets were used to develop an empirical relationship between lead-dust loading and lead-dust concentration (EPA 1998; Rasmussen et al 2013; Adgate et al 1995; Lanphear et al 1995; Clayton et al 1999). The summary statistics for the five data sets are presented in Tables 4 and 5.

Table 4: Summary Statistics for Lead-Dust Loading (µg/ft^2) for Studies used to Develop the Loading to Concentration Relationship

Study	Sample size	Range (Min, Max)	AM	GM
HUD 1990 raw	312	0.51 to 375	19.48	7.55
HUD 1990 clean	301	0.51 to 375	19.30	7.52
Adgate 1995 raw	444	0.37 to 10,219.3	not reported	20.2
Adgate 1995 clean	394	0.37 to 10,219.3	82.8	19.9
CHDS 2013 raw	1022	0.019 to 89.43	2.88	0.88
CHDS clean	1010	0.042 to 89.43	2.92	0.94
Rochester raw	204	1.15 to 2,114.2	130.0	49.78
Rochester clean	190	1.37 to 1,993.53	160.59	57.86
NHEXAS raw	244	0.051 to 105,816.6	527.1	5.49
NHEXAS clean	215	0.2 to 9,792	64.1	6.5

Table 5: Summary Statistics for Lead-Dust Concentration (µg/g) for Studies used to Develop the Loading to Concentration Relationship

Study	Sample size	Range	AM	GM
HUD 1990 raw	312	0.09 to 50,400	521.89	189.27
HUD 1990 clean	301	15.8 to 6,320	364.21	197.06
Adgate 1995 raw	444	19 to 33,000	not reported	490
Adgate 1995 clean	394	19 to 33,000	902.2	494.3
CHDS 2013 raw	1022	14.2 to 7,800	210.5	119.1
CHDS clean	1010	14.2 to 7,800	212.2	119.9
Rochester raw	204	12.2 to 4,233.9	593.9	361.7
Rochester clean	190	20.56 to 7,069.04	906.29	486.67
NHEXAS raw	244	0.928 to 30,580	469.8	114.04
NHEXAS clean	215	13.99 to 30,580	489.9	185.42

Comparison of Metrics using Canadian House Dust Study

The CHDS data is unique in that it was the only study that measured, rather than estimated, all three lead-dust metrics for a large national surveillance monitoring study. The CHDS data was analyzed in several different ways which supports the positive correlation between lead-dust loading and lead-dust concentration, the log-log linear shape of this relationship, and correlation between different sampling approaches which measure or estimate the same lead-dust metric (lead-dust loading or lead-dust concentration).

Whole-house lead-dust loading values were estimated using a vacuum method and a wipe method. Central tendency values based on room-by room wipe data were estimated in different ways as shown in Table 6. Across all approaches, there was a positive correlation between the lead-dust loading values reported between wipe and vacuum sampling approaches. The weighted average arithmetic and geometric mean accounts for higher lead-dust loadings reported in entryways compared to the rest of the house. Weighting measurements by surface-area sampled when multiple samples are available is a common technique (AHHS 2011). In this instance the surface area covered by each room was not available so 10% was assigned to the entryway and 90% was assigned to the rest-of-home samples.

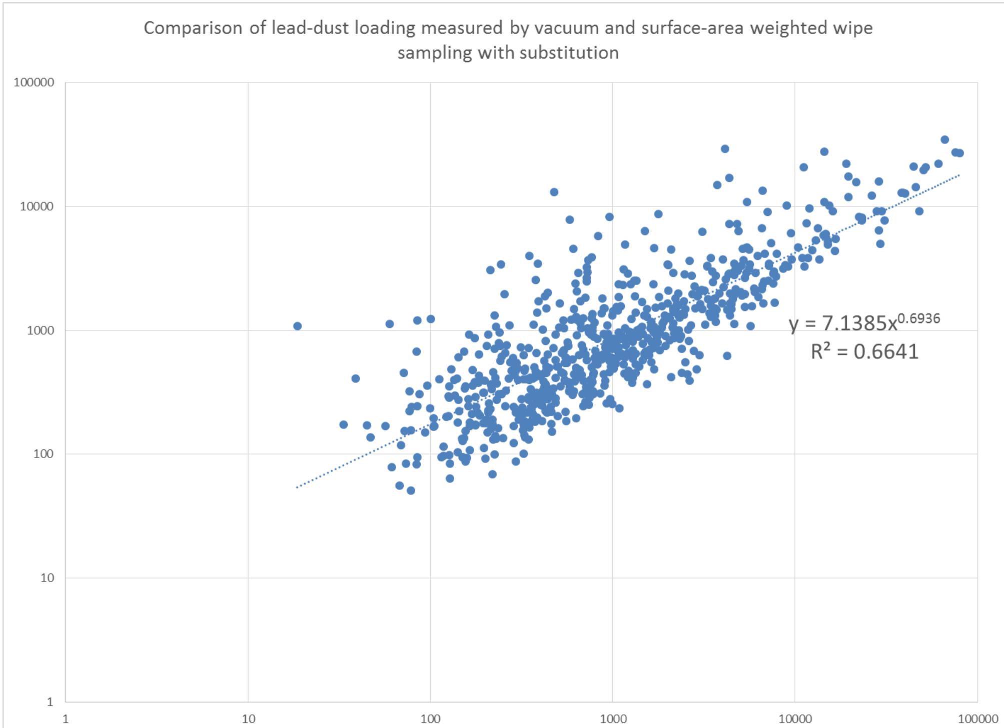
HUD recommends that at least four rooms per house are sampled (HUD 2012). For samples with less than four wipes present per house, the vacuum whole-house lead-dust loading values was substituted for the missing wipe value. Whole-house lead-dust loading values are available and a reasonable surrogate for missing wipe data because these estimates represent average lead-dust loading across all carpeted and hard surface flooring within the residence, while wipe samples represent lead-dust loading on hard surfaces.

Table 6: Results of Regression analysis between (Ln)Lead-dust Loading for wipe and vacuum sampling method

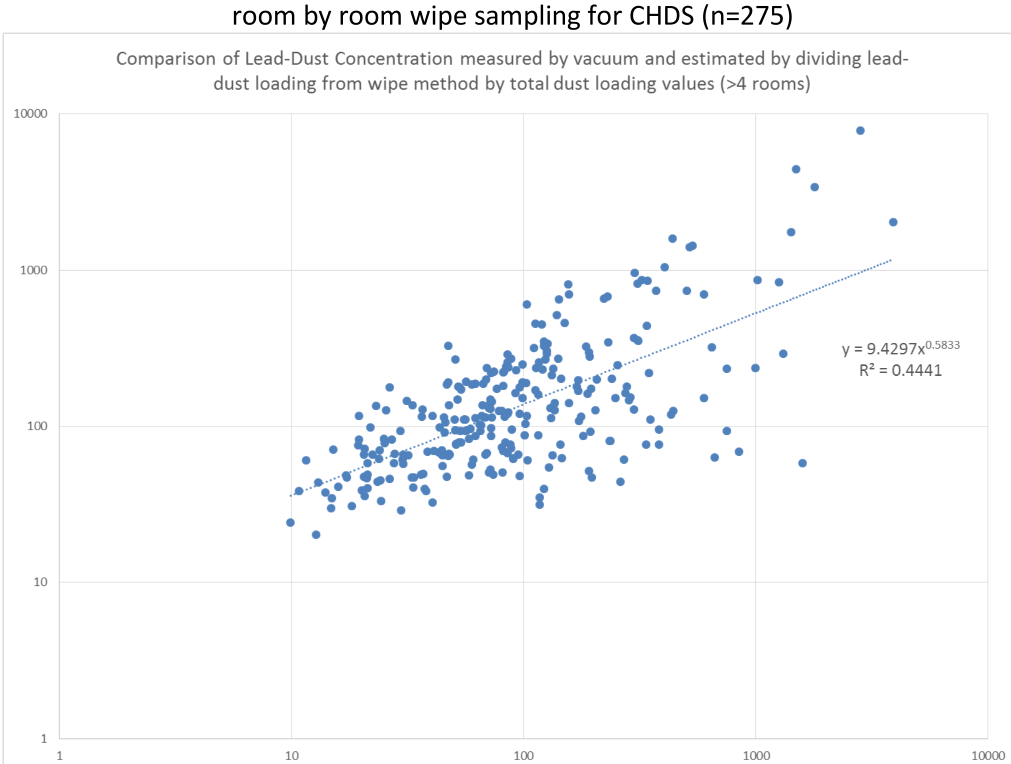
How central tendency wipe values were derived for comparison with vacuum lead-dust loading values	R^2 All Rooms n=700	R^2 <4 rooms n=425	R^2 4+ rooms N=275
1) Arithmetic Mean	0.23	0.23	0.27
2) Geometric Mean	0.24	0.28	0.28
3) Weighted average Arithmetic mean	0.24	0.23	0.29
4) Weighted average Geometric mean	0.24	0.24	0.30
5) Weighted average Arithmetic mean with substitution	0.66	0.67	0.69
6) Weighted average Geometric mean with substitution	0.51	0.59	0.48

Figure 1 uses the measured lead-dust loading data obtained through the room by room wipe sampling (combination five) and compares this with estimated lead-dust loading values obtained through the vacuum protocol. Figure 2 uses the measured lead-dust concentration data obtained through the vacuum sampling protocol and compares this with an estimated lead-dust concentration. The estimated lead-dust concentration was calculated by dividing the weighted average arithmetic mean lead-dust loading (µg/ft^2) values by their matched total-dust loading (g/ft^2) value. This was done for homes with four or more rooms samples. This is similar to the approach taken by Hunt et al., to estimate dust-lead concentration solely with wipe sampling (Hunt et al., 2012).

<u>Figure 1:</u> Comparison of Lead-Dust Loading (µg/ft^2) data measured by whole-house vacuum and room by room wipe sampling for CHDS (n=700)



<u>Figure 2:</u> Comparison of Lead-Dust Concentration (µg/g) data measured by whole house vacuum and room by room wipe sampling for CHDS (n=275)



Results of the Lead Loading to Concentration Relationship

The loading to concentration regression analysis was run for each raw data set, each cleaned dataset, all raw pooled data, and different pooled combinations of the cleaned datasets. For each regression, a slope, standard error of slope, intercept, standard error of intercept, overall standard error of the regression, and R^2 were derived. The average ln(loading) values was also calculated. All of these values are used to estimate lead-dust concentration values from lead-dust loadings, and can be used in loading to concentration calculator (Supplemental File 1).

The overall results for individual raw data sets and cleaned data sets show consistency across data sets. As shown in Table 7, the standard error of the regression was reduced and the R^2 was increased after data cleaning procedures (elimination and substitution of data points) were employed. Cleaning the data sets also reduced the differences between data sets to some degree.

Table 7: Results of Lead-Dust Loading to Lead-Dust Concentration Regression for Individual raw and cleaned Data Sets

Data Source	**(1a) CHDS-raw/ (1b) CHDS-clean**	**(2a) HUD-raw/ (2b) HUD-clean**	**(3a) Adgate-raw/ (3b)- Adgate clean**	**(4a) Rochester raw/ (4b) Rochester clean**	**(5a) NHEXAS-raw/ (5b)- NHEXAS clean**
Sample Size	1022/1010	312/301	312/394	202/190	244/215
Slope	0.399/0.424	0.656/0.608	0.469/0.442	0.434/0.457	0.634/0.565
Standard error of slope	0.014/0.015	0.039/0.031	0.029/0.026	0.046/0.043	0.036/0.035
Intercept	4.780/4.787	5.243/5.283	6.240/6.204	6.07/6.187	4.736/5.226
Standard error of Intercept	0.020/0.020	0.050/0.039	0.044/0.036	0.065/0.0654	0.079/0.050
Standard Error	0.662/0.659	0.896/0.693	0.778/0.733	0.928/0.897	1.237/0.741
ln(loading) avg	-0.125/-0.058	2.022/2.018	3.067/3.008	3.889/4.058	1.644/1.869
R^2	0.41/0.42	0.47/0.55	0.44/0.42	0.30/0.37	0.55/0.55

When individual data sets were pooled together, the standard error of the regression was reduced and R^2 was increased, compared to individual data sets. As shown in Table 8, the pooled data sets increase the degrees of freedom and stabilize the slope estimates. The slope drives the incremental increase in lead-dust concentration values from observed lead-dust loading values.

Table 8: Results of Loading to Concentration Regression for Pooled, Cleaned Data sets

Data Source	**(1) CHDS, HUD, Adgate Baseline**	**(2) CHDS, HUD, Adgate, clean**	**(3) CHDS, HUD, Adgate, NHEXAS**	**(4) CHDS, HUD, Adgate, Rochester**	**(5) CHDS, HUD, Adgate, NHEXAS, Rochester**	**All data sets raw**
Sample Size	1643	1705	1920	1895	2110	2174
Residual Degrees of Freedom	1641	1703	1918	1893	2108	2172
Slope	0.421	0.438	0.437	0.416	0.420	0.425
Standard error of slope	0.009	0.009	0.008	0.008	0.008	0.009
Intercept	5.142	5.202	5.204	5.301	5.293	5.219
Standard error of Intercept	0.018	0.017	0.016	0.016	0.016	0.019
Standard Error	0.737	0.705	0.723	0.735	0.744	0.889
ln (Loading)	0.881	1.017	1.113	1.322	1.378	1.323
R^2	0.55	0.58	0.56	0.58	0.57	0.50

All pooled regression results show consistency in the estimated central tendency lead-dust concentrations across all combinations. The percent difference between estimated lead-dust concentrations range from 0.9% to 11.8% for lead-dust loading values ≤100 µg/ft^2. The percent difference between estimated lead-dust concentrations range from 4.3% to 18.8% for lead-dust loading values >100 µg/ft^2. Estimated central tendency lead-dust concentrations for a wide range of lead-dust loading values are provided in Table 9.

Table 9: Estimated Central Tendency Lead-Dust Concentration values across Loading to Concentration Regression combinations

Lead-Dust Loading (µg/ft^2)	**(1) CHDS, HUD, Adgate Baseline**	**(2) CHDS, HUD, Adgate, clean**	**(3) CHDS, HUD, Adgate, NHEXAS**	**(4) CHDS, HUD, Adgate, Rochester**	**(5) CHDS, HUD, Adgate, NHEXAS, Rochester**
0.1	44.8	42.4	40.9	44.4	42.4
1.0	118.0	116.3	111.9	115.7	111.5
10	311.2	319.0	306.1	301.5	293.3
100	820.5	874.5	837.2	785.9	771.6
1,000	2,163.0	2,397.4	2,290.0	2,048.1	2,029.4
10,000	5,702.5	6,572.7	6,263.8	5,337.7	5,337.9

Taken together, the overall results show a consistent slope and overall relationship suggesting that there are underlying mechanistic reasons across studies for why the relationship between lead-dust loading and lead-dust concentration continues to present as log-log linear. Figures 3 and 4 compare the baseline loading to concentration relationship (combination one) and the loading to concentration relationship using all cleaned data (combination 5). Combination five reflects all data cleaning and pooling steps described in the methods. All combinations from Table 8 show similar scatterplots as presented for Figures 3 and 4.

Figure 3: Scatterplot of the Baseline Loading to Concentration Relationship (Combination One)

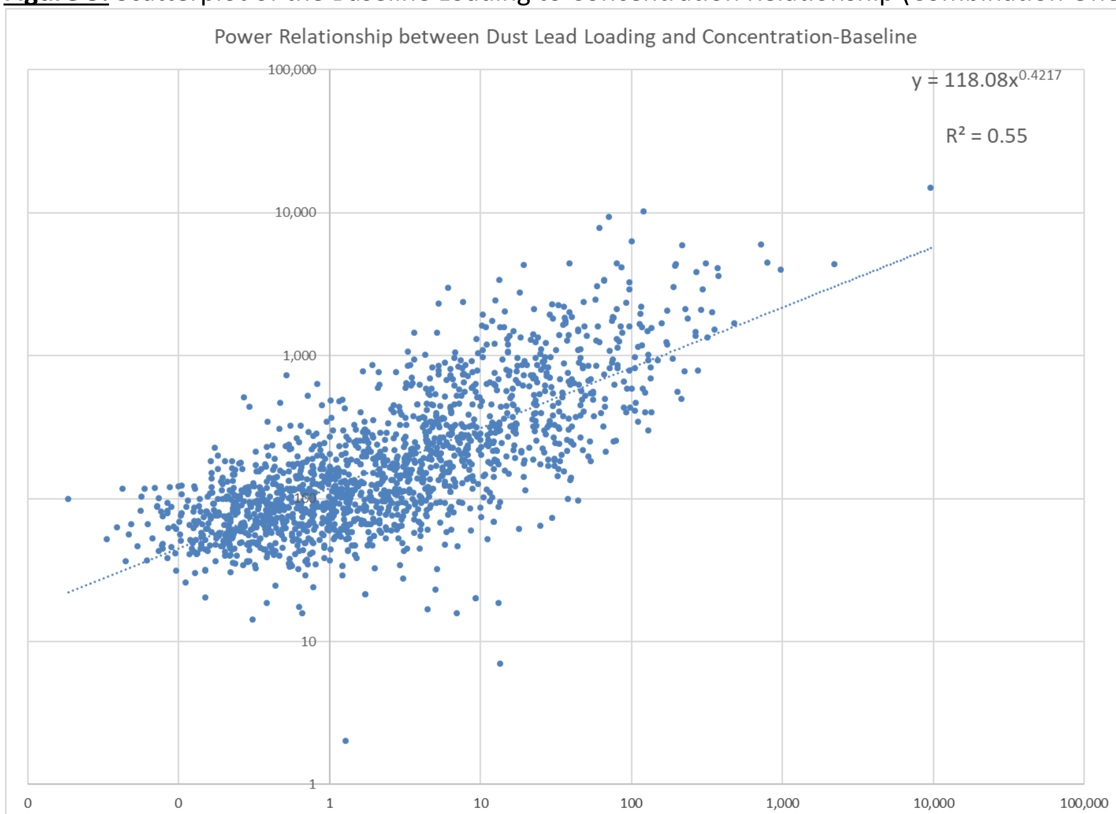
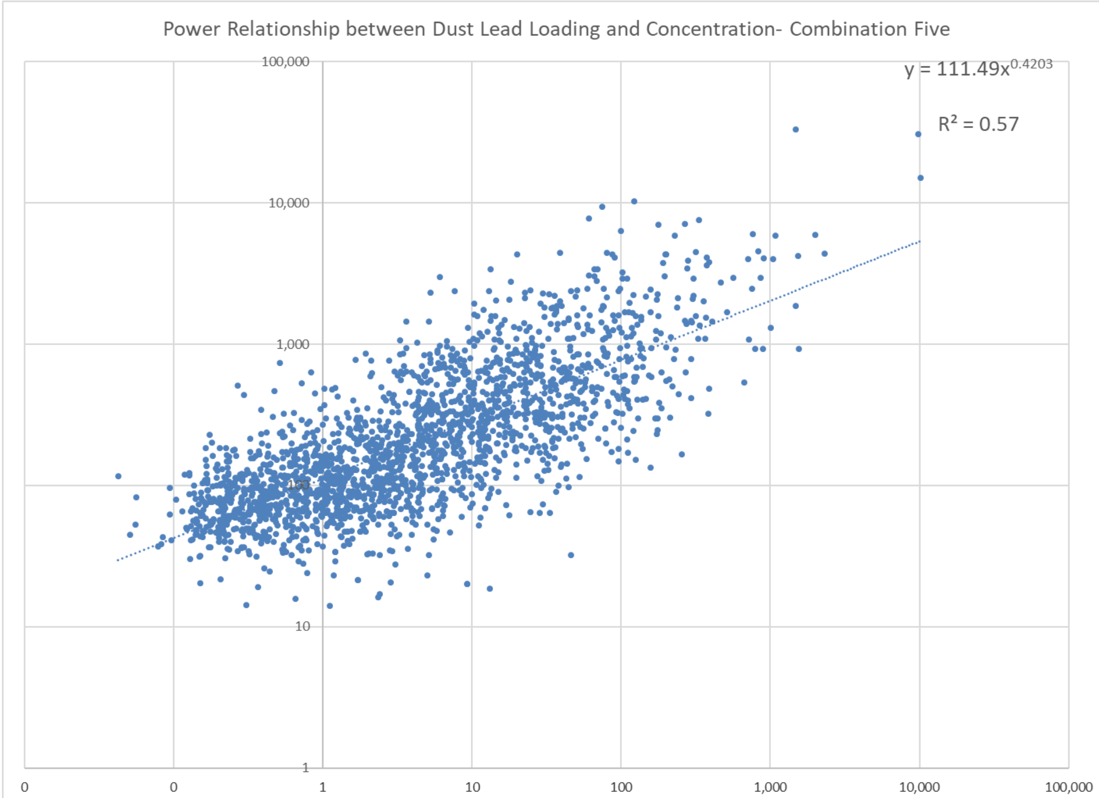


Figure 4: Scatterplot of the Pooled, Cleaned Loading to Concentration Relationship, Combination Five



Summary of Extracted Data from Previous Lead-Dust Monitoring Studies

The purpose of this literature search was to identify additional published studies which reported both lead-dust loading and lead-dust concentration, and therefore could be used for evaluation of the loading to concentration regression developed in the present analysis. Extraction of the data for this purpose, resulted in ancillary observations about study types and study design. These observations are included in Supplemental File 2. As summarized in Table 10 and Figure 5, studies were assigned into four groups: national, regional, intervention, and historically contaminated.

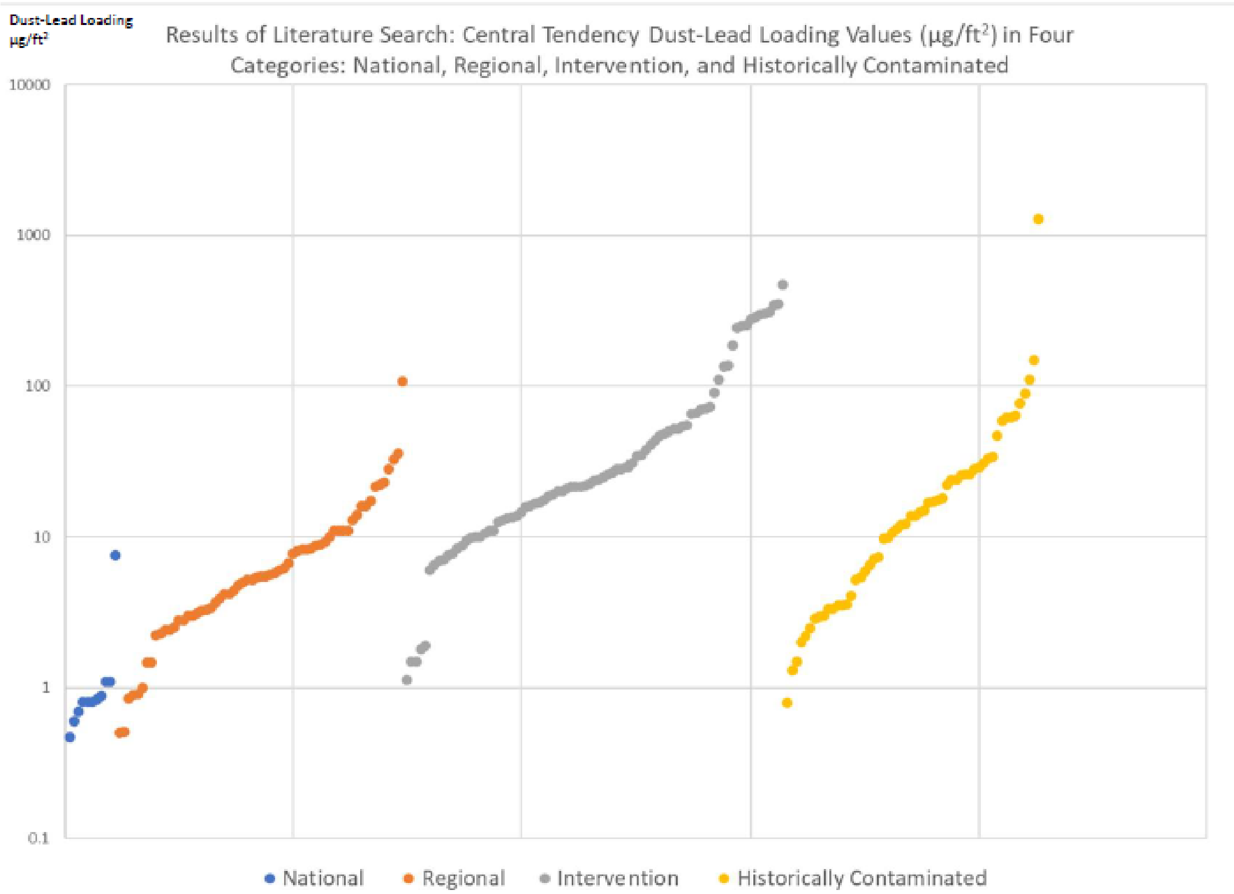
Table 10: Percent of unique sampling groups for studies that reported lead-dust loading with central tendency estimates in various lead-dust loading categories

Study Type	Number of Groups	<1 µg/ft²	between 1 and 10 µg/ft²	CT between 10 and 40 µg/ft²	CT >40 µg/ft²
National	10	72.8%	27.2%	-	-
Regional	44	7.9%	66.7%	23.8%	1.6%
Intervention	42	-	18.1%	45.8%	36.1%
Historically Contaminated	32	1.8%	39.3%	41.1%	17.9%

The full distribution of studies with larger sample sizes at the national and regional scale likely overlap with and contain some of the more highly elevated groups in intervention and historically contaminated areas. However, large-scale surveillance studies are not designed to characterize exposures to these more highly exposed groups.

The loading to concentration relationship error variance term estimates larger variability for lead-dust concentrations as lead-dust loading values increase. The variability is smaller with lower lead-dust loadings. This is an important consideration when interpreting lead-dust loading monitoring results at central tendency or upper percentile values, especially when those results are used in an empirical regression to estimate lead-dust concentration. This overview of lead-dust loading values provides context for general trends associated with central-tendency lead-dust loading values reported across four broad study types.

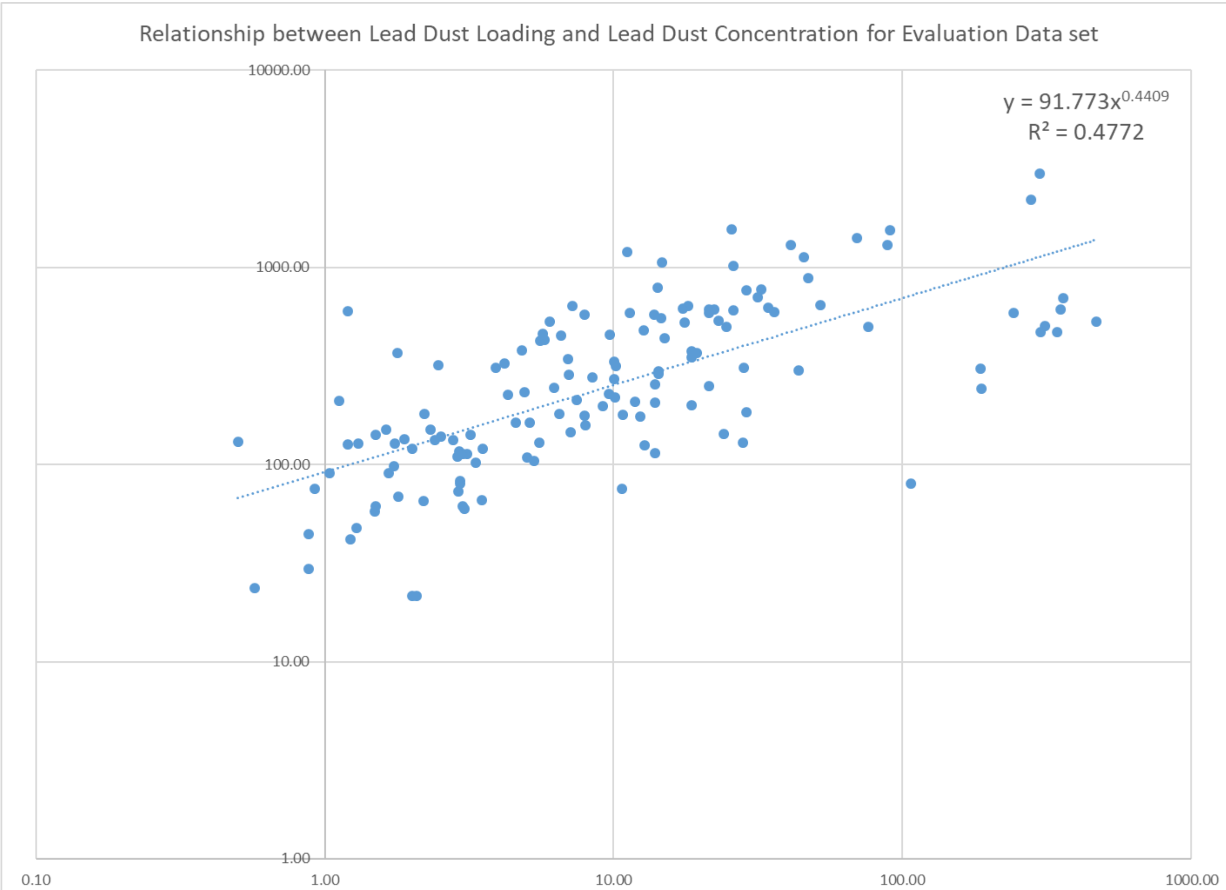
<u>Figure 5:</u> Central Tendency Lead-Dust Loading values reported across different study types (one dot for each unique sampling group within each study)



Evaluation of Lead Loading to Concentration Relationship

Paired central-tendency lead-dust loading and lead-dust concentration values from independent data sources were compiled. There were thirty-two studies identified with both lead-dust loading and lead-dust concentration reported. Within each study, more than one sampling group was identified. When possible, summary statistics from unique sampling groups were used. There were 142 data points identified that had matched lead-dust loading, lead-dust concentration central tendency values. The summary data from these studies shows consistency with the raw data sets used to derive the loading to concentration relationship. As shown in Figure 6, the data are positively correlated and the scatterplot indicates the same log-log linear shape, with lead-dust concentration increasing more slowly as lead-dust loading increases.

Figure 6: Scatterplot of Paired Central Tendency Dust-Lead Loading and Dust-Lead Concentration Data from the Literature



A meta-analysis looking at the variability surrounding the slopes, intercepts, and overall standard error across these individual studies and/or unique sampling groups was beyond the scope of this analysis, but could be undertaken in the future. A descriptive observation suggests that the regression outputs (slope, intercept, standard error) of central tendency data points from the literature are within the range of the regression outputs for data sets used to develop the loading to concentration regression.

Other descriptive observations used to evaluate the loading to concentration regressions are: (1) consideration of different prediction intervals, and (2) the percent difference between median and mean values in the evaluation data set. The prediction interval estimates the probability that an independent dust-lead concentration value will lie within the prediction interval for a given dust-lead loading value. In its update of the lead-dust hazard standard, EPA used a prediction interval of 50% (EPA 2019). Table 10 shows that a prediction interval of 80% increases the percent of measured lead-dust concentration values that fall within the predicted range. The percent difference between the median and mean measured and predicted value across the 142 data points suggests that the loading to concentration regression reasonably predicts central-tendency lead-dust concentration values.

Table 10: Results of Comparison of Predicted and Observed Lead Dust Concentration values from the Literature

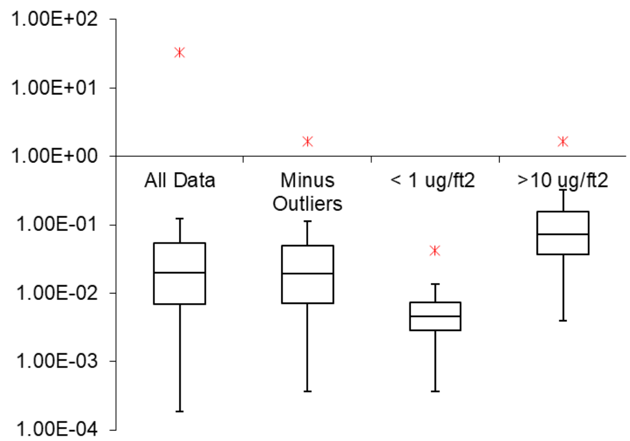
	Percent of Data Points within 50% Prediction Interval Range (25/75)	Percent of Data Points within 80% Prediction Interval Range (10/90)	Percent Difference between median observed and estimated concentration (n=142)	Percent Difference between average observed and estimated concentration (n=142)
#1- CHDS, HUD, Adgate Original	77.5%	91.5%	16.3%	-1.2%
#2- CHDS, HUD, Adgate Clean	75.4%	90.8%	18.1%	3.1%
#3- CHDS, HUD, Adgate, NHEXAS	77.5%	91.5%	14.7%	-1.1%
#4- CHDS, HUD, Adgate, Rochester	76.1%	91.5%	13.6%	-5.0%
#5- CHDS, HUD, Adgate, NHEXAS, Rochester	76.8%	92.3%	11.2%	-7.4%

Relationship between Lead Loading and Total Dust Loading

Lead loading and total dust loading are correlated. Background lead-dust loading levels are generally at or below 1 µ*g/ft*^{2} (Lanphear et al., 1998; HUD 2011; EPA 2019). Elevated lead-dust loading levels are generally defined as above 10 µ*g/ft*^{2} (McDonald et al., 2010; Rasmussen et al., 2013; EPA 2019). As total dust loading increases, lead-loading also tends to increase. The empirical data indicate that elevated lead-dust loading levels (>10 µ*g/ft*^{2}) are unlikely when total dust loading levels are low. It is also unlikely that total-dust loading levels will be elevated when lead-dust loading values are low (<1 µ*g/ft*^{2}). Many studies have reported this relationship for national and regional studies (Sutton et al., 1995; Hunt et al., 2012) (Whitehead et al., 2015). Some studies with highly elevated dust-lead levels in intervention or historically contaminated sites have also reported this relationship while others have not (Meyer et al., 1999; Succop et al., 1998). When site-specific data are available, professionals may find it useful to enter their own site-specific lead-dust loading and lead-dust concentration monitoring data into the calculator found in Supplemental File 1.

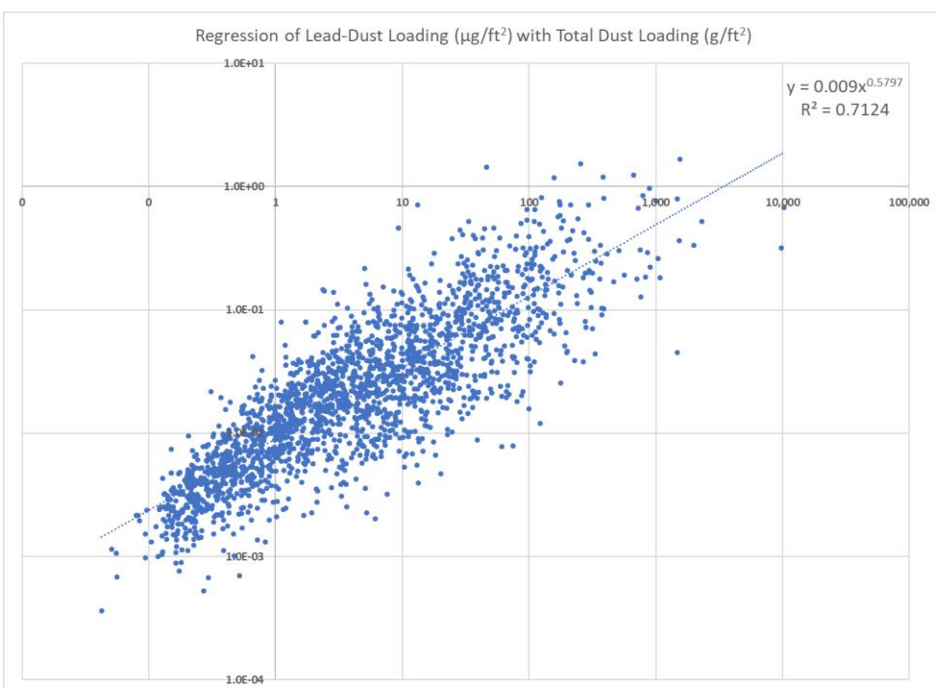
For the studies with raw data considered in the present analysis, there is a clear difference between the mean values for total dust-loading values (g/ft^2) matched with background and elevated lead-dust loading values (µg/ft^2).

Figure 7: Box-plot of Total Dust-Loading values across categories: (1) all data, (2) all data minus outliers, (3) for when lead-dust loading is <1 µ*g/ft*^{2}, and (4) for when lead-dust loading was >10 µ*g/ft*^{2}.



While figure 7 shows that it is unlikely that elevated dust-lead loading values are associated with low total dust loading values and vice-versa, Figure 8 suggests that there is an underlying relationship between dust-lead loading and total dust loading that is even stronger than the relationship between lead-dust loading and lead-dust concentration.

Figure 8: Scatterplot of the relationship between lead-dust loading (µg/ft^2) and total dust loading (g/ft^2) for all data minus outliers



This relationship suggests use of constant values for total-dust lead loading or fixed ratios between lead-dust concentration and lead-dust loading are not a good-fit for conversion between lead-dust loading and lead-dust concentration. The alternative presented in this analysis directly compares lead-dust loading and lead-dust concentration. The most likely reason why one lead-dust loading value is associated with a range of lead-dust concentrations is because of the variability associated with total dust loading.

Discussion

There are limitations to this study which contribute to uncertainty. For analysis of raw data, assumptions were made when contextual data was not present. For example, it was not possible to digitize all the raw data points from the CLEARS study. Therefore, some data points are missing (Adgate et al., 1995) (EPA 2019). The Rochester study did not report the surface area present in each room, and the number of rooms sampled within a residence did not match for one-third of the samples. For this analysis, an assumption was made that 10% of surface area was covered by the entryway and 90% was covered by other rooms. The same assumption was made for the CHDS data which reported total surface area sampled, but not surface area present within each room. This could be further explored through later sensitivity analyses or research to determine which distribution improves fit of the data.

The evaluation of the loading to concentration regression with extracted data was limited by the data available in extracted studies. Some, but not all of the 32 studies from the literature contained more detailed summary statistics with both measures of central tendency, measures of dispersion (standard deviation, geometric standard deviation), or percentiles, which could have allowed for more detailed meta analyses. For studies where information is available, further evaluation of the loading to concentration evaluation through meta-analysis could be explored.

Another limitation across all studies arises from the use of different sampling and analytical approaches. This contributes to potential measurement error which is likely greater for the oldest studies. This could be further explored as new data sets become available by replacing older data sets with newer data sets that adhere to current sampling and analytical guides and standards.

There are many related variables that affect lead-dust concentration, other than lead-dust loading. Examples of influencing factors include the cleaning frequency, floor type, condition of interior paint, size and lead concentration of ambient air particles. It is notable that a relatively simple empirically based lead-dust loading/lead-dust concentration regression has a significant slope and explains over half the variance in the natural logarithm of the data. The log-log form of the relationship also has a physical basis, in that the distribution of lead-dust concentrations arises from random multiple dilutions of dust from multiple sources (EPA 2019).

The empirical relationship between lead-dust loading and lead-dust concentration developed from this data is based on log-transformation of both metrics. Changes in lead-dust loading tend toward greater increases in lead-dust concentrations at lower loadings compared with higher loadings. The log-log linear relationship between lead dust loading and lead dust concentration is based in empirical evidence. However, the primary mechanistic reason for this relationship is the variable total dust loading. If total dust loading was constant across all homes, then the average lead dust concentration would be linear with respect to the lead dust loading.

Some studies have attempted to use a single dust-loading value or a fixed ratio to describe this relationship (Gulson et al., 2018; LeBot et al., 2010; Glorennec et al., 2012). However, many empirical data sets where total dust loading have been measured show that this is not the case (Rasmussen et al 2013) (Sutton et al 1995). Like most monitoring data, total dust loading is lognormally distributed and highly variable. The slope between lead dust concentration and lead dust loading increases at a slower rate at higher loadings. In order for lead-dust concentration to increase at a slower rate as lead-dust loading increases, the total dust loading must also increase. This is most likely because lead-dust loading and total dust loading are correlated as shown in Figure 8.

Lead-dust loading and total dust loading could be correlated because homes with more lead also have poor flooring conditions which make dust clean-up more difficult. They could also be correlated because of lower cleaning frequencies, more carpeted surfaces, more occupants per household, presences of smokers, or presence of pets. Another rationale could be that lead is principally from a baseline source of dust while homes with higher quantities of total dust have additional dust sources without lead (e.g., pet dander, non-contaminated soils, etc.) which dilute the baseline lead concentrations. Efficient and frequent removal of dust through cleaning is an effective strategy to reduce total dust-loading and associated lead-dust loading values. Several intervention studies have shown that reduction of total dust-loading through cleaning also reduces lead-dust loading. However, the empirical data does not fully explain why the total dust loading is correlated with the lead-dust loading and for what reasons (EPA 2019).

As new lead-dust monitoring studies are designed and implemented, researchers are encouraged to report out multiple lead-dust metrics in any of the following ways:

- Report total dust loading by recording the total mass of dust collected and surface area sampled using a wipe or vacuum sampling method, AND
- Report lead-dust loading using wipe samples for at least four rooms within a home or through composite wipe sampling OR report lead-dust loading through vacuum samples on both hard flooring and carpeted surfaces, AND
- Report lead-dust concentration through vacuum samples of both hard flooring and carpeted surfaces.

HUD Guidance recommends sampling at least four rooms within a home when wipe samples are collected (HUD 2012). Further, some rooms appear to be more predictive of whole-house lead-dust loading values than others. Common choices include the living room, the room where the child spends the most time such as a children's bedroom, kitchen, and entryway. Whole house estimates of lead-dust loading based on one, two, or three wipe samples are likely to either under or overestimate the actual whole house lead-dust loading depending on where the sample was collected. Most studies recommend using a combination of rooms with hard surface flooring and carpeting (HUD 2012).

When vacuum sampling methods are used, sampling a larger surface area provides a more representative sample within the home. HUD Guidance recommends composite sampling for flooring of the same type for wipe sampling (HUD 2012). No such recommendation exists for vacuum sampling. Nevertheless, many studies have consistently shown that carpeted surfaces tend to have higher total dust loading values compared to hard floors. Recording the surface area and flooring type sampled can provide important context when interpreting lead-dust monitoring data.

Conclusion

The goal of this study is to evaluate overall trends associated with lead-dust loading, lead-dust concentration, and total-lead loading when raw data are available. Empirical statistical relationships were explored using different combinations of raw data sets. Subsequently these regressions were evaluated using additional studies from the literature. This analysis also shows that substitution and elimination of individual data-points slightly improves, but does not change the baseline statistical relationship between lead-dust loading and lead-dust concentration. Central tendency results from a literature search showed consistency between the observed concentrations and concentrations that were estimated using various loading to concentration regressions. General consistency across study-types was observed despite differences in study-design, location, and timing. This consistency suggests an underlying physical basis for the observed log-log linear relationship between lead-dust loading and lead-dust concentration, through indirect influence of total-dust loading. When only lead-dust loading values are present, this empirical statistical relationship provides a reasonable approach to estimate dust lead concentrations.

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