Environmental Pollution 229 (2017) 290–299

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Geospatial evaluation of lead bioaccessibility and distribution for site specific prediction of threshold limits☆

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ARTICLE INFO

Article history:
Received 31 January 2017
Received in revised form
18 May 2017
Accepted 23 May 2017
Available online 7 June 2017

Keywords:
Lead
Bioaccessibility
Contaminant modeling

ABSTRACT

Recent work identified the need for site-specific Pb bioaccessibility evaluation and scaled contaminant modeling. Pb heterogeneity has made bioaccessibility characterization difficult, and complicated distribution models. Using field testing, bioaccessibility measurement, Integrated Exposure Uptake and Biokinetic (IEUBK) modeling, and geospatial techniques, we propose a framework for conducting applied risk-based, multiscale assessment. This framework was tested and implemented in Burlington, VT, an area of old housing stock and high Pb burden (up to 15 000 mg kg^{-1}) derived primarily from paint. After analyzing local soil samples for total and bioaccessible Pb, it was determined that bioaccessible and total Pb were well correlated in this area, through which an average bioaccessibility parameter was derived approximating Pb bioaccessibility for this soil type and Pb impact. This parameter was used with the IEUBK to recommend the local limit for residential soil Pb be reduced from 400 to 360 mg kg^{-1}, taking into consideration the lowering of the blood lead level threshold for Pb poisoning from 10 to 5 μg dL^{-1} by the Centers for Disease Control (CDC). Geospatial investigation incorporated samples collected during this investigation and samples from a high school summer science academy, and relied on three techniques, used at different scales: kriging of total and background Pb alone, kriging of total and background Pb with housing age as a well-sampled, well-correlated secondary variable (cokriging), and inverse distance weighting of total and bioaccessible Pb. Modeling at different scales allowed for characterization of Pb impact at single sites as well as citywide. Model maps show positive correlation between areas of older housing and areas of high Pb burden, as well as potential at different scales for reducing the effects of Pb heterogeneity.

1. Introduction

Since pre-industrial times, Pb has been used within commercial and residential products, resulting in surficial enrichment worldwide and establishing it as a primary marker of the Anthropocene (Marx et al., 2016). During the 20th century, Pb was ubiquitous, appearing within solder used on food cans as well as within gasoline and paint (Laidlaw and Filippelli, 2008). Pb is a persistent neurotoxin, and sustained contact poses acute developmental risk to children and fertility risks to adults (ATSDR, 2007). Soil and dust sources constitute the main pathway of Pb transmission to children (Walraven et al., 2015). Pb contamination is concentrated in urban centers where historical patterns of suburbanization and deindustrialization and contributions overlap render residents most vulnerable to legacy pollutants (McClintock, 2015). Bioavailability of Pb in soil can vary widely according to speciation, soil characteristics, and across sites affected by different Pb sources (Davis et al., 1993; Hettiarachchi and Pierzynski, 2004; Ruby et al., 1999), contributing to frustrate attempts at creating a unified bioavailability model across soil types (Zhu et al., 2016). As a result the establishment of a single threshold for Pb contamination in soils is arbitrary and impossible (Henry et al., 2015; Palmer et al., 2015), as it may overestimate or underestimate bioavailability. Instead, bioavailability and distribution of soil Pb may be locally investigated and assessed for the establishment of individualized Pb contamination threshold, a method that can inform land use practices and aid in the assessment of lead hazard citywide.

Pb possesses a high residence time in soils, where it is tightly held by functional groups on soil organic matter in most natural environments (Kaste et al., 2006; Schroth et al., 2008). Pb behavior in soils is complex, with reactivity and fate governed by soil

☆ This paper has been recommended for acceptance by Dr. Yong Sik Ok.

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http://dx.doi.org/10.1016/j.envpol.2017.05.064
0269-7491/

properties, equilibrium reactions and biogeochemical processes. These factors affect whether Pb occurs as a free hydrated ion, within inner-sphere complexes with mineral surfaces, complexed with organic matter functional groups, or coprecipitated with inorganic molecules (Reeder et al., 2006). Pb species possess varying solubility and reactivity, which, along with particle size and availability of reactive sites, dictate the extent to which soil Pb is bioavailable (Traina and Laperche, 1999). Extraction-based *in vitro* procedures can artificially measure Pb bioaccessibility, which can be used to derive relative bioavailability to estimate Pb bioavailability in natural systems (Drexler and Brattin, 2007; Ruby et al., 1999).

Mielke and others (Laidlaw et al., 2016; Mielke et al., 2013) established a spatial correlation between Pb in city centers and elevated blood Pb levels (BLL). Through exploratory XRF field testing, it was determined that Burlington possesses high soil Pb due to a high proportion of older housing containing historical Pb paint. This study targets areas of highest contamination by sampling soil adjacent to painted structures, comparing this data with samples collected further from structures approximating non-paint impacted soil Pb levels (>2 m). This approach was developed to address the need for targeted sampling, as more traditional gridded methods of sampling citywide may fail to detect high Pb concentrations near structures. Using this data, Pb concentration can be estimated at unsampled sites through interpolative geospatial and geostatistical models, including kriging, cokriging, and inverse distance weighing (IDW) (Cattle et al., 2002; Stewart et al., 2014; Moukana et al., 2013). Schwarz et al. (2012), identified the importance of scale in modeling Pb distribution, as larger Pb particles from structures tend to be localized, resulting in elevated soil Pb adjacent to structures, while background levels tend to be lower and comprised of finer particles. Such contrast between spatially delimited influences points to the need for incorporating different modeling scales, which we have expanded to include different modeling techniques. Kriging, which describes a range of least-squares regression algorithms, is used to evaluate the distribution of a given attribute in unsampled areas by averaging measured values in the same neighborhood (Goovaerts, 1999). If data is highly skewed, as can occur when an attribute has a low background level but wide range of values, transformations are appropriate before subjecting the data to modeling (Wu et al., 2006). When existing data is not comprehensive enough to form an accurate prediction, a well-correlated, comprehensively sampled secondary variable may be added to improve the estimation of the model, a technique known as cokriging. Like kriging, IDW is built on the assumption that an attribute at a single location is a weighted average of known data points, with the difference that in IDW, closer data points are given greater weight than data points farther away (Goovaerts, 1999; Robinson and Metternicht, 2006). With the help of these methods, remediation efforts can be expanded from areas with high reported BLLs and high Pb in soil and dust to include areas of high predicted BLLs and soil Pb identified by modeling. Bioaccessibility parameters and Pb concentration in soil, dust, and water can then be used to extrapolate the expected burden of Pb to children using biokinetic modeling (Laidlaw and Taylor, 2011; Stewart et al., 2014; White and Marcus, 1998).

The goal of this study is to use targeted sampling to establish a framework for examining Pb paint-impacted soil hazard in a city with old housing stock (Burlington, VT) for use in preventive health efforts and site remediation. Once Pb bioavailability and distribution were constrained for a single area, this data was used to develop a local, site-specific safe soil Pb threshold, and estimate citywide risk using predictive blood lead level modeling and geostatistical interpolation techniques.

2. Materials and methods

2.1. Study location

The city of Burlington (VT), with over 47% of houses built prior to 1950, was chosen as a study site. Most these older homes consist of wooden frame structures covered by clapboards painted with multiple generations of paint, including older Pb paint layers. Burlington soils are comprised primarily of a single soil type (Windsor loamy sand), comprised of glaciofluvial sediment. A local Housing and Urban Development (HUD) funded program, the Burlington Lead Program (BLP), part of the City of Burlington Community and Economic Development Office, provides lead hazard outreach and abatement assistance to the community in the form of soil barriers and window replacement, and served as a community partner in this investigation. According to available data from the BLP (data.burlingtonvt.gov), the mean construction date of houses chosen for remediation is 1906 ($n = 325$), suggesting paint is an important local contributor of Pb. The Old North End and South End neighborhoods contain the highest concentration of sites receiving window and soil barrier remediation treatments from the BLP (SI Fig. 1). Though paint is considered the principal source of Pb within this area, the possibility of minor tetraethyl lead (TEL) contributions from pre-catalytic gasoline should be acknowledged. In particular, the washing of wind deposited particulate TEL from structures into the surrounding soil, indicates that urban soil adjacent to exterior walls may contain multiple sources of Pb (Mielke et al., 2011). For modeling purposes, and given Burlington's history of low traffic density, we have chosen to focus on the relationship between older housing and Pb, but measurements may reflect more than one source.

2.2. Sample collection and preparation

One hundred and one soil samples were collected between 0 and 10 cm at 9 public and residential sites in and around Burlington, during the summer and fall months of 2015 and spring months of 2016. Sample locations are given in Fig. 2a and b. Residential sites were sampled with permission. Samples were augured from within 0.5 m of building exteriors, from all accessible sides, with control samples taken at least 2 m away from buildings at each site. If mulch or grass was present, sample was obtained from soil underlying groundcover. After sampling, visible organic material was separated, samples were dry sieved through 2 mm mesh, and a fraction was ball-milled for 7 min using a steel ball mill.

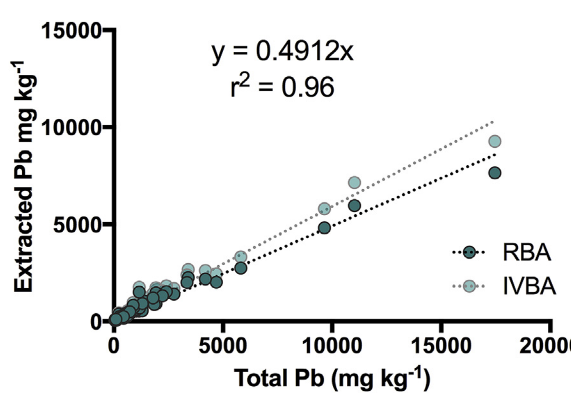

Fig. 1. Extractable (bioaccessible) vs. total Pb within contaminated Burlington soils. Transparent symbols represent *in vitro* bioavailable Pb (IVBA), non-adjusted; opaque symbols represent relative bioavailability, adjusted from IVBA using weighted linear regression. Regression formula and r^2 value represent RBA values only.