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**MEMORANDUM**

**OLEM Directive 9200.1-128**

**SUBJECT:** Recommendations for Sieving Soil and Dust Samples at Lead Sites for Assessment of Incidental Ingestion

**FROM:** *for* Dana Stalcup, Director *Schatz Fitz-James*  
Assessment and Remediation Division  
Office of Superfund Remediation and Technology Innovation

**TO:** Superfund National Program Managers, Regions 1 - 10

The purpose of this memorandum is to transmit the Technical Review Workgroup for Metals and Asbestos (TRW) Technical document entitled "Recommendations for Sieving Soil and Dust Samples at Lead Sites for Assessment of Incidental Ingestion." This document contains a review of the current data on the relationship between the particle size fractions sieved at lead-contaminated sites and the likelihood that they will adhere to hands and be incidentally ingested.

Based on their review, the TRW recommends moving from the current < 250  $\mu\text{m}$  particle size to < 150  $\mu\text{m}$  particle size. The recommendation is based upon an expanding body of evidence illustrating that dermally adhered soil is dominated by particle fractions < 150  $\mu\text{m}$ . The weight of evidence is sufficiently strong to update the recommended sieving size while the impact on the Integrated Exposure Uptake Biokinetic Model (IEUBK) is assessed and the standard operation procedure for the *in vitro* bioavailability assay (IVBA) is updated. In the interim, the TRW Lead Committee recommends that the particle size fraction used for soil lead concentration in the fine fraction be the same as the particle size fraction used for the determination of site-specific bioavailability using the IVBA, and used for determining site-specific background.

This report and other efforts related to addressing lead in soil can be found on the Internet at <https://www.epa.gov/superfund/lead-superfund-sites-technical-assistance>. Please contact Michele Burgess at [Burgess.Michele@epa.gov](mailto:Burgess.Michele@epa.gov) or (703) 603-9003 if you have questions or concerns.

Attachment

1. "Recommendations for Sieving Soil and Dust Samples at Lead Sites for Assessment of Incidental Ingestion"

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**RECOMMENDATIONS FOR SIEVING SOIL AND DUST SAMPLES AT LEAD SITES FOR ASSESSMENT OF INCIDENTAL INGESTION****OVERVIEW**

The purpose of this document is to review the data currently available on the relationship between the particle sizes, dermal adherence and lead enrichment, as well as to revise the definitions of coarse and fine fractions for soil and dust for use in lead risk assessment. This document provides the technical basis for updating the recommended particle size fractions sieved at lead-contaminated sites. The intended audience for this document is human health risk assessors who are familiar with sieving soil and dust samples for use in risk assessments. For further background information on sampling procedures in lead risk assessment, refer to U.S. Environmental Protection Agency (2013, 2007a, 2003) or the Technical Review Workgroup for Lead (TRW) website (<https://www.epa.gov/superfund/lead-superfund-sites>).

Since 2000, the Office of Land and Emergency Management (OLEM) has recommended sieving dry (<10% moisture) soil and dust samples through a No. 4 (4.75 mm) or a No. 10 (2.0 mm) sieve (ASTM, 1999) to remove any large debris (*e.g.*, sticks, stones; U.S. EPA, 2000). The resulting material, referred to as the “total soil sample”, is then weighed and sieved through a No. 60 sieve to produce the “coarse” (>250  $\mu\text{m}$ ) and the “fine” (<250  $\mu\text{m}$ ) fractions. This fine fraction is intended to represent a reasonable upper-bound estimate of the soil and dust fraction that is most likely to stick to hands (or other objects that a child may put it its mouth) and be subsequently ingested (U.S. EPA, 2007a, 2000). In addition, the concentration of lead in the <250  $\mu\text{m}$  particle size fraction was used to calibrate the Integrated Exposure Uptake Biokinetic Model for Lead in Children (IEUBK model; U.S. EPA, 2000, 1994), and in the development of the *in vivo* and *in vitro* bioavailability assays for lead in soil (Smith et al., 2011; U.S. EPA, 2009, 2007b; Casteel et al., 1997; Maddaloni et al., 1998; Ruby et al., 1996).<sup>1</sup>

U.S. EPA performed a literature search for relevant data on the relationship between particle size and dermal adherence, and between particle size and lead enrichment (January 2000-December 2011). Based on more recent information, the TRW now recommends that dry total samples (as defined above) be weighed and sieved using a No. 100 W.S. Tyler® sieve<sup>2</sup>, or equivalent, to identify the “coarse” (>150  $\mu\text{m}$ ) and the “fine” (<150  $\mu\text{m}$ ) fractions for use in the assessment of human health risks for soil and dust exposures (see Appendix A for further sampling information). This recommendation is based on a growing body of evidence showing that dermally-adhered soil and dust, representative of soil and dust exposure to young children via incidental ingestion, is dominated by particles <150  $\mu\text{m}$  (see Tables 1 and 3, Figure 1). In addition, the more recent information also indicates the potential for enrichment of lead in

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<sup>1</sup> The <250  $\mu\text{m}$  particle size fraction was used for the development of the default lead bioavailability value and recommended for use in laboratory analyses to develop site-specific lead bioavailability values.

<sup>2</sup> Mention of specific products or manufacturers should not be interpreted as an endorsement.

smaller sized particles at some sites (see Tables 2 and 4). The TRW recognizes, however, that this recommendation to sieve to 150  $\mu\text{m}$  to obtain the fine fraction may be fractionated further (*e.g.*, 125, 50, 10  $\mu\text{m}$ ) or larger particle size fractions considered as site-specific history or circumstances warrant.<sup>3</sup>

## **INTRODUCTION**

Incidental ingestion is the primary pathway for childhood exposure to lead in soil and dust (with the exception of pica behavior) and is governed by dermal adherence (Ruby and Lowney, 2012; Lanphear et al. 2002; Lanphear et al. 1998; Succop et al. 1998; Landrigan et al. 1975).<sup>4</sup> Blood lead (PbB) levels associated with incidental ingestion are related to the concentration of lead in soils in the vicinity of and dust on surfaces of a child's home, as well as lead adhered to hands of infants and the hands of their parents (Ruby and Lowney, 2012; Simon et al., 2007; Laidlaw et al., 2005; NRC, 2005; Mielke and Reagan, 1998; Succop et al. 1998; Lanphear and Roghmann, 1997; Landrigan et al. 1975). Links between lead in environmental media and blood samples have also been corroborated with statistical models predictive of PbB and isotopic studies which "follow the lead" from yards and house dust to the hands and blood of children (Gulson et al., 2009; Manton et al., 2000; Angle et al., 1995). Lead-contaminated house dust typically includes a soil component, and is a strong predictor of PbB levels in children (NRC, 2005; Lanphear et al., 2002, 1998).

Site-specific risk assessment requires that soil and dust samples accurately represent incidentally ingested material. Soil particle size, an important determinant of dermal adherence, is inversely associated with contaminant concentration, mobility, and bioavailability (Beamer et al., 2012; Madrid et al., 2008; ATSDR, 2007; Madrid et al., 2002; Lanphear et al., 2002; Manton et al., 2000; Lanphear et al., 1998; Sterling et al., 1998). As such, reliable data on the particle size fraction that is most likely to adhere to children's hands and on the lead concentration found in that particle size can improve the accuracy of exposure and risk calculations in lead risk assessments. The TRW Lead Committee previously determined that the lead concentration in the <250  $\mu\text{m}$  particle size fraction is more representative of ingestion exposure than unsieved soil (U.S. EPA, 2000). While this remains true, more recent studies identified by the TRW Lead Committee were reviewed and the findings support using the <150  $\mu\text{m}$  particle size fraction, as it is more representative of exposure to lead via incidental ingestion of soil and dust.

### **Dermal Adherence is Dominated by Fine Particles**

The TRW identified ten studies reporting dermal adherence of soil and dust as a function of particle size fractions (*e.g.*, <150, <125, <40  $\mu\text{m}$ ) from a variety of sample types (*e.g.*, shooting ranges, mining sites, urban environments). See Tables 1 and 3; also Figure 1. These studies

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<sup>3</sup>Representative site-specific data are essential for developing a risk assessment (as well as cleanup goals) that reflect the current or potential future conditions. Ultimately, lead exposure is determined by the ingested dose ( $\mu\text{g Pb/kg-body mass/day}$ ). For this report, the mass of ingested lead ( $\mu\text{g/Pb}$ ) is intended to equal to the concentration of lead in the contaminated media multiplied by the mass of this media that is ingested.

<sup>4</sup> It is known that some individuals deliberately ingest soil (pica) and that these individuals may have soil ingestion rates well in excess of the typical ingestion levels used in most U.S. EPA risk assessments. Pica exposure is generally not assessed in Superfund lead risk assessments.

indicated that dry (<10% moisture) soil and dust particles in the <150 µm fraction were more likely to adhere to hands than larger fractions (Gong et al., 2013; Bergstrom et al., 2011; Juhasz et al., 2011; Siciliano et al., 2009; Choate et al., 2006a; Yamamoto et al., 2006; U.S. EPA, 2000, 1995; Sheppard and Evenden, 1994; Duggan et al., 1985; Que Hee et al., 1985; Duggan, 1983). In general, adherence generally increases with decreasing particle size (Choate et al., 2006a; Driver et al., 1989). As shown in Figure 1, approximately 90% of the cumulative mass of soil adhered to children's hands falls within the <150 µm fine fraction. Smaller particles are more mobile than larger fractions and are more likely to accumulate in the indoor environment, as a result of deposition of wind-blown soil or track-in transport of soil on clothes, shoes, pets, toys, and other objects, providing additional opportunity for exposure to this particle size fraction (Luo et al., 2011; Schmidt, 2010; Layton and Beamer, 2009; Laidlaw and Filippelli, 2008; Qian et al., 2008; Bright et al., 2006; Laidlaw et al., 2005; Gulson et al., 1995). See Tables 1 and 3.

### **Contaminant Concentration Typically Increases as Particle Size Decreases**

The TRW identified 19 studies concerning lead concentrations in soil and dust for different particle size fractions (see Tables 2 and 4). Together these studies indicated that enrichment in concentration for smaller particle size fractions is dependent upon site-specific characteristics, and enrichment in smaller particle sizes may not occur at all sites. However, particle size distribution of metals in shooting ranges, incinerators, mine tailings and associated background soil samples from three mining sites, as well as urban soils and dusts demonstrated consistent enrichment in particle size fractions smaller than <150 µm (Kim et al., 2011; Luo et al., 2011; Juhasz et al., 2011; Madrid et al., 2008; Pye et al., 2007; Ljung et al., 2007, 2006; Weiss et al., 2006; Momani, 2006; Tawinteung et al., 2005).

### **RECOMMENDATIONS FOR SIEVING AT LEAD CONTAMINATED SITES**

Based on this analysis, the TRW generally recommends that “total” samples (as defined as the total of dust and soil on page 1) be weighed and sieved through a No. 100 W.S. Tyler® sieve<sup>5</sup> or equivalent to identify the “coarse” (>150 µm) and the “fine” (<150 µm) fractions for use in the assessment of human health risks for soil and dust exposures to lead (see Appendix A for further sampling information). The fine fraction (<150 µm) has increased potential for incidental ingestion based on stronger relative dermal adherence, an increased likelihood to accumulate in the indoor environment (through deposition of wind-blown soil and/or transport track-in of soil on clothes, shoes, pets, toys, and other objects), and the likelihood of enrichment of lead in smaller particle size.

On a site-specific basis, it may be appropriate for risk management decisions to consider the benefits of obtaining information on the other particle size fractions (*e.g.*, <250, <125, <63, <50 µm) to better relate ingestible size fractions with site history or site-specific conditions.<sup>6</sup> For example, it may be appropriate to consider larger particle size fractions at some sites such

<sup>5</sup>See ASTM E11 and ISO 565 for more information.

<sup>6</sup> Representative site-specific data are essential for developing a risk assessment (as well as cleanup goals) that reflect the current or potential future conditions. Ultimately, lead exposure is determined by the ingested dose (µg Pb/kg-body mass/day). For this report, the mass of ingested lead (µg/Pb) is intended to equal to the concentration of lead in the contaminated media multiplied by the mass of this media that is ingested.

as firing ranges (where lead bullet fragments may exist) or sites where wet soil contact may be expected (larger particles can adhere to hands when wet). Soil and house dust samples in the <175 µm fraction collected from the Bunker Hill Superfund Site in Kellogg, ID have been used in the IEUBK model (after an initial calibration) to accurately and consistently predict PbB levels in the community for more than 15 years (von Lindern et al., 2003; PHD, 1986; Snee, 1982; Yankel et al., 1977). Alternatively, smaller particle size fractions may also be informative for certain sites. At this time, the TRW Lead Committee does not have specific recommendations for alternative particle size intervals for soil and dust. Users may contact the TRW Lead Committee to discuss site-specific conditions that may warrant consideration of alternative particle size fractions<sup>7</sup>.

This recommendation is consistent with U.S. EPA recommendations for particulate sampling under RCRA (U.S. EPA, 2002). In addition, particulate sampling theory recognizes that sampling errors are reduced when smaller particles are sampled (Barcan et al., 1998; Gy, 1998, 1992, 1982). To promote defensible and reproducible site investigations and decision making, while maintaining flexibility needed to respond to different site conditions, EPA recommends the Data Quality Objectives process (U.S. EPA, 2006). Data Quality Objectives provide a structured approach to collecting environmental data that will be sufficient to support decision-making: <http://www.epa.gov/QUALITY/dqos.html>.

Currently, these recommendations specifically apply to lead risk assessment, but the importance of particle size as it relates to dermal adherence, consequent ingestion, and variance in contaminant levels may also apply to other metals, polyaromatic hydrocarbons, or other contaminants in soil and dust (Ruby and Lowney, 2012; Beamer et al., 2012; Bergstrom et al., 2011; Siciliano et al., 2009; Yamamoto et al., 2006).

The TRW Lead Committee recognizes that the recommendation to sieve soil samples to a particle size fraction representing <150 µm differs from previous recommendations and also differs from the particle size used for validation of the IEUBK model and the *in vitro* bioaccessibility assay for lead (IVBA) (U.S. EPA, 2009, 2007b). However, the weight of evidence is sufficiently strong to update the recommended sieving size while the impact on the IEUBK model is assessed and Standard Operating Procedure for the IVBA is updated. In the interim, the TRW Lead Committee recommends that the particle size fraction used for soil lead concentration in the fine fraction be the same as the particle size fraction used for the determination of site-specific bioavailability using the IVBA and use for determining site-specific background.

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<sup>7</sup> <https://www.epa.gov/superfund/lead-superfund-sites-guidance>

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Table 1. Summary of soil adherence studies.\*

Study	Year	Data Reporting Method	Study Type and Conditions	Particle Size Cutoff Accounting for 50% of Adhering Mass	Particle Size Cutoff Accounting for 90% of Adhering Mass
Bergstrom et al.	2011	Estimated mass adhering for each size fraction	Laboratory	NR	NR
Siciliano et al.	2009	Mass of soil adhering for each size fraction	Field Laboratory: agricultural soils Laboratory: Brownfield soil	40 $\mu\text{m}$ 40 $\mu\text{m}$ 125 $\mu\text{m}$	130 $\mu\text{m}$ 370 $\mu\text{m}$ 760 $\mu\text{m}$
Choate et al.	2006	Mass of soil adhering for each size fraction	Laboratory: low moisture soil Laboratory: medium moisture soil Laboratory: high moisture soil	33 $\mu\text{m}$ 44 $\mu\text{m}$ 80 $\mu\text{m}$	110 $\mu\text{m}$ 120 $\mu\text{m}$ 220 $\mu\text{m}$
Yamamoto et al.	2006	Mass of soil adhering for each size bin	Field	67 $\mu\text{m}^{\text{d}}$	134 $\mu\text{m}^{\text{d}}$
Kissel et al.	1996	Mass of soil adhering for each size fraction	Laboratory: dry soil Laboratory: wet soil	62 $\mu\text{m}$ 150 $\mu\text{m}$	210 $\mu\text{m}$ 350 $\mu\text{m}$
Sheppard and Evenden	1994	Enrichment ratios of the mass of specific size fractions adhering	Laboratory	NR	NR
Driver et al.	1989	Mass of soil adhering for each size fraction	Laboratory	NR <sup>c</sup>	NR
Duggan and Inskip	1985	Mass of soil adhering for each size fraction	Laboratory	57 $\mu\text{m}$	130 $\mu\text{m}$
Duggan et al.	1985	Number of particles adhering in each size range	Field	NM <sup>a</sup>	NM
Que Hee et al.	1985	Mass of house dust adhering for each size fraction	Laboratory	NA <sup>b</sup>	NA

<sup>a</sup>NM = No mass-based estimate of soil adherence. 90% of particles were <10  $\mu\text{m}$ .

<sup>b</sup>NA = Not applicable (study used house dust not soil).

<sup>c</sup>NR = Not reported or not calculable from data presented.

<sup>d</sup>Average value for the population of children (three of nine) with the largest soil particles adhering.

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Table 2. Summary of soil enrichment studies.

Study	Year	Sample Type	Particle Size Cutoff for Pb Enrichment	Comment
Gong et al.	2013	Arable soils	<53 $\mu\text{m}$	The distribution of most heavy metals in different size particles was increased with decreasing particle size. The mass loading of heavy metals in micro-aggregates was high while their leachability was very low.
Beamer et al.	2012	Residential	<63 $\mu\text{m}$	Outdoor soil had an increased Pb concentration at <63 $\mu\text{m}$ .
Juhasz et al.	2011	Shooting range	<50 $\mu\text{m}$	<50 $\mu\text{m}$ fraction, the average concentration of lead was approximately twice the average concentration of lead in the <2 mm fraction but varied between 0.86 - 5.31. Lead enrichment in the <50 $\mu\text{m}$ particle size fraction was up to 5 times the concentration observed in the bulk soil.
Kim et al.	2011	Mining site	<125 $\mu\text{m}$	Increased Pb concentration with decreased particle size.
Lou et al.	2011	Urban	<10 $\mu\text{m}$	2-10 $\mu\text{m}$ had increased metal concentrations; 50-280 $\mu\text{m}$ was less marked.
Smith et al.	2011	Shooting range, incinerator, fill, mining, gas work	<250 $\mu\text{m}$	Pb concentration in the <250 $\mu\text{m}$ fraction was greater than the <2mm fraction.
Acosta et al.	2009	Urban soils	<75 $\mu\text{m}$	Pb concentration in the <75 $\mu\text{m}$ fraction was consistently higher in two of four parks sampled. This was attributed to vehicular depositions.
Siciliano et al.	2009	Agricultural, Brownfield	<45 $\mu\text{m}$	Increasing metal concentrations with decreasing particle size fractions.
Ajmone-Marsan et al.	2008	Urban	<10 $\mu\text{m}$	$\geq 50\%$ of the total Pb accumulated in the <10 $\mu\text{m}$ fraction.
Madrid et al.	2008	Urban	<50 $\mu\text{m}$	Particle sizes <50 $\mu\text{m}$ had generally higher lead concentrations than >50 $\mu\text{m}$
Ljung et al.	2007	Urban	<50 $\mu\text{m}$	Lead concentrations were consistently higher in <50 $\mu\text{m}$ fraction
Pye et al.	2007	Near River	<150 $\mu\text{m}$	Mean and maximum Pb concentrations increased with decreasing particle size fraction
Ljung et al.	2006	Urban	<50 $\mu\text{m}$	Demonstrated average 1.5 times enrichment for metals in finest fraction (<50 $\mu\text{m}$ ) compared with either the bulk sample or the 50-100 $\mu\text{m}$ fraction.
Momani	2006	Urban	<63 $\mu\text{m}$	The greatest Pb concentrations were in the <63 $\mu\text{m}$ fraction.
Weiss et al.	2006	Urban	<63 $\mu\text{m}$	84% of the particles were in the range of 125–500 $\mu\text{m}$ . The highest concentration of lead was in the smallest fraction analyzed (<63 $\mu\text{m}$ ).
Tawinteung et al.	2005	Battery recycling plant, lead smelting factory	<150 $\mu\text{m}$	Pb concentration was ~2 times higher in the <150 $\mu\text{m}$ than in the 250 $\mu\text{m}$ -2mm fraction.
Young et al.	2002	Industrial facilities, roadways	<38 $\mu\text{m}$	Pb in PM <sub>10</sub> was enriched by 1.10 to 8.31times compared with bulk soil samples. Pb in fine soil (<38 $\mu\text{m}$ ) was enriched by 1.12 to 7.83 times compared with coarse soil samples (300-2000 $\mu\text{m}$ ).
Al-Raijhi et al.	1996	Urban	<20 $\mu\text{m}$	Pb in the 20 $\mu\text{m}$ size fraction was enriched 1.5- to 3.0 times compared with the 250 $\mu\text{m}$ size, and 2.5- to 6.4 times compared with the 1500 $\mu\text{m}$ size fraction.



Gulson et al.	1995	Mining, residential	<150 $\mu\text{m}$ (soil) <100 $\mu\text{m}$ (dust)	Finer fractions contain 2-9 times higher concentration than bulk fractions.
Kitsa et al.	1992	Chromium Contaminated Site	<38 $\mu\text{m}$	Increased Pb concentrations with decreased particle size.

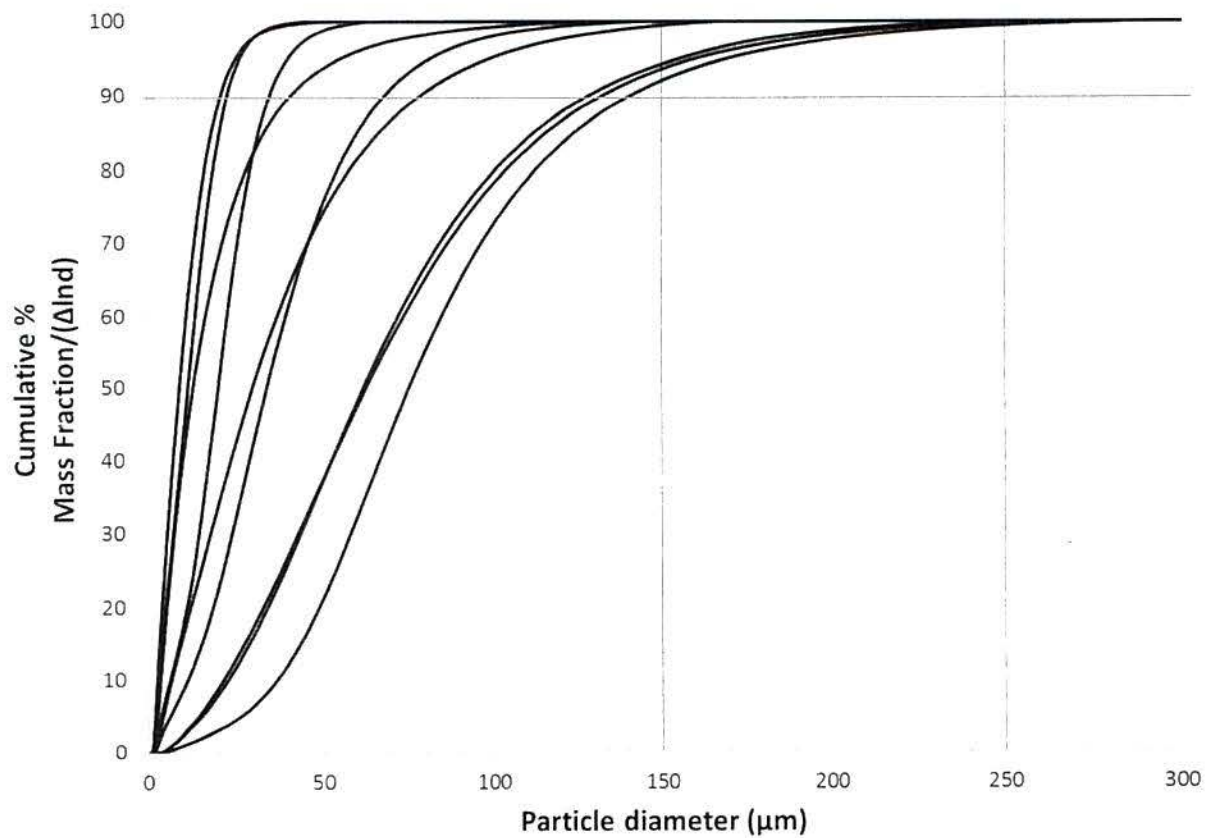


Figure 1. Mass fraction of soil adhering to children's hands as a function of particle size (data from Yamamoto et al., 2006). The curves represent the cumulative mass fraction adhering as a function of particle size for 9 individual children. The published figure was provided courtesy of Michael V. Ruby and adapted above with permission from Ruby and Lowney (2012). Copyright 2012 American Chemical Society.

Table 3. Summary of available particle adherence studies.

Site-Specific Information	Subjects	Sampling Procedure	Particle Size Tested (µm) Moisture Content (% Moisture)	Comments	Reference
Mining and smelting sites, quarry – river banks and slag  (Coeur d' Alene River Basin, ID)	Adult volunteers (3M, 3F)	Hand washing method with soil  Dry and wet conditions	<63 63-150 150-250 250-2000  Wet: 3.5-14.7%; Dry: <0.25%	Wet media always adhered to a greater extent than dry media. Adhered media generally had higher elemental concentrations than bulk media. Over 60% of the adhered fraction was <63 µm; this is particularly interesting for the Black Sand Beach (BSB) because the data show approximately 97% of the BSB material is in the 250 µm - 2mm fraction. While most metals concentrations increased with decreasing particle size, this paper includes some data that show the highest concentration in the bulk soil (e.g., slag material), and others that show little trend with particle size. Regression analyses suggest smaller particle fractions may have higher elemental concentrations. Results of application of a maximum likelihood estimation technique generally indicate that handling of dry media leads to preferential adherence of smaller particle sizes, while handling of wet media does not. Because adhered material can differ greatly in particle size distribution from that found in bulk material, use of bulk concentrations in exposure calculations may lead to poor estimation of actual exposures.	Bergstrom et al., 2011
Clay loam (CSU, Colorado);  Silty clay loam (ISU, Iowa)	Adult volunteers (n=108)	Unwashed hands;  hand press technique – soil removed with distilled water or adhesive tape  Also evaluated washing hands prior to exposure	<25 25-38 38-63 63-125 125-250 250-500 500-2000  Wet: 3.35-10.1% Dry: <2%	This study measured the effect of soil moisture, particle size distribution (PSD) of soil and organic carbon content on the particle sizes that adhere to human skin. Adhered fraction consisted primarily: <ul style="list-style-type: none"> <li>• &lt;63 µm for dry and 'moderately moist' soils (&lt;3.81 %)</li> <li>• &lt;250 µm for wet soils (&gt;3.81%)</li> </ul> No estimates of potential ingestion were made in this analysis. Also investigated the effect of commonly used method of removing the adhered soil mass (by washing with water) on the disaggregation of soil particles (which would bias the measurement of adhered particle size towards the smaller particle sizes). The authors are interested in dermal exposure rather than incidental ingestion; therefore, they are interested in studying how long soil sticks to skin because that dictates how much of the contaminant transfers into the skin (they cite EPA 2001).  This paper does not include data on adherence or data on the concentration as a function of soil/sediment particle size; however, the paper assesses the potential bias introduced when water is used to remove the adhered material from the skin (water tends to disaggregate some soil particles). The authors developed an algorithm for estimating the PSD of a (dry) from the results of wet-sieving the soil. Two soil types were included: clay loam and silty loam. The data indicate disaggregation was not significant for the silt loam for the particles sizes less than 500 µm but it was significant for the clay loam. This paper is useful for the discussion of methodology issues & limitations.	Choate et al., 2006a  Choate et al., 2006b
Five soil types (Virginia, US) (11 soils samples)	Adult Males	Hand press; pre-post contact weights were measured  Dry-sieved	≤150 ≤250 Unsieved soil  moisture content not measured;	Summarized in EPA 1995: Researchers examined soil adherence to skin by particle size. Increasing mass and size of the larger particles may reduce the total soil mass that adheres.  <150 increased adherence more than <250 µm: An average of 0.6, 0.9, and 1.4 mg of soil for the unsieved fractions, <250, and <150, respectively, adhered to each square centimeter of skin on hands (mg/cm <sup>2</sup> ).  Results showed that an obvious conclusion from the data is that finer soil particles adhere more readily to hands than do coarser particles. The most important factor to determine soil adherence (based on particle size, soil type, and organic content) was particle size, followed by soil type. Used to support 250 µm recommendation in 2000 TRW Soil Sampling Guidance.	Driver et al., 1989

Site-Specific Information	Subjects	Sampling Procedure	Particle Size Tested ( $\mu\text{m}$ ) Moisture Content (% Moisture)	Comments	Reference
Street dust collected from gutters or pavement of main and side roads  (London, England)	NR	"taking a pinch of dust" between the thumb and forefinger and rubbing the digits together such that the surplus falls off; weight difference used to estimate adherence	<500	Authors reported that "a number of tests with several different people" gave a range of 2 to 7 mg or a mean of about 4 mg dust retained per finger and thumb, or 2 mg per digit. No other particle sizes were tested.	Duggan & Williams, 1977
Review of the literature	NA	NA	NA	Retrospective analysis using NHANES and UK PbB data, plus extensive literature review. In attempt to explain observed discrepancy in child vs. adult exposure, propose that dust lead levels correlate with PbB levels. Used to support 250 $\mu\text{m}$ recommendation in 2000 TRW Soil Sampling Guidance.	Duggan, 1983
Schoolyard dusts  (London, UK)	Children (n=368; 5-6 yrs old)	Hand wipe, hand wash  "dry conditions"	<180  <500 (bulk - dried and ground until it was sieved to 180 $\mu\text{m}$ )	90-98% of particles were less than 10 $\mu\text{m}$ ; largest particles were 100-180 $\mu\text{m}$ (as cited by US EPA, 1995)  Used to support 250 $\mu\text{m}$ recommendation in 2000 TRW Soil Sampling Guidance.	Duggan et al., 1985
Activity-based soil samples (soccer/rugby field, farmers, groundskeepers, kids in mud, taekwon do, greenhouse)	Adults, Children  (n=101: 92 subjects outdoor; 9 subjects indoor)	Skin surface wash (legs, hands, forearms, faces, feet)	Varied wet and dry conditions	Soil loading study that did not differentiate between specific particle sizes, but are activity dependent.	Kissel et al., 1996a
Five soil samples  (Washington State, US)  (sand, loamy sand, sandy loam, silt loam)	1 adult female	Hand press; measured dermal soil loading and adherence  Clay and organic carbon contents were determined	$\leq 150$  150-250  $\geq 250$  Wet: (12-18%)  Dry: (<2%)	Discusses that for wet samples, >150 $\mu\text{m}$ may be appropriate, but for dry samples, "mass adherence is predominantly attributable to sub-150 $\mu\text{m}$ , and perhaps even sub-65 $\mu\text{m}$ particles." Under dry conditions adherence varied inversely with grain size <150, 150-250, >250 $\mu\text{m}$ fractions. In general much greater soil loading with wet samples, in some cases finest fraction had less adherence when wet.	Kissel et al., 1996b

Site-Specific Information	Subjects	Sampling Procedure	Particle Size Tested ( $\mu\text{m}$ ) Moisture Content (% Moisture)	Comments	Reference
Soils (Cincinnati, OH)  Dust (residential)	Adults (n=10);  Children (n=6; 3-10 yrs old)	Hand wipes  Dry sieved and ground until it passed through a 149 $\mu\text{m}$ sieve	<149  Subset of house dust (1 house)  <44 44-149 149-177 177-246 246-392 392-833	Found no relationship between particle size and skin adherence of dry house dust in very limited measurements following artificial loading. Assumptions were made – soil is composed of particles of the indicated diameters, all soil types and particle sizes adhere to the skin, and an equivalent weight of particles of any diameter adhere to the same surface area of skin.  Adherence of house dust particles to hands, for fractions less than 246 $\mu\text{m}$ , no particle size difference in dermal adhesion. Concluded that loose dust particles less than 246 $\mu\text{m}$ , sieved from dust collected in their study houses, would be more likely to adhere to a child's hands than would larger particles and, therefore, would be more likely to be ingested by the child  The particle size-specific adherence results were produced using a single "small adult."	Que Hee et al., 1985
Garden soils  (Ontario, Manitoba Canada enriched with Pb)	1 subject	Hand press/ soil crumbling/ hand wash  Dry sieved	<50 <41 <114 114-500  Dry	The largest enrichment of the investigated contaminants was associated with sandy soils, where the bulk of the adsorbed contaminants were found on the finer particles that adhere readily to skin but make up only a small fraction of the total soil mass. Lower soil loads on the skin showed greater enrichment, with a marked increase at soil loads below levels that would call for obvious washing of the hands.  Clay particles (<2 $\mu\text{m}$ ) fit in size with the roughness characteristics of the skin, implying that the finest particle sizes may be resistant to the washing of hands, but soil particles <50 $\mu\text{m}$ in diameter tended to preferentially adhere to dry skin, regardless of soil type. Adsorbed soil constituents (Pb, Hg, I, U and C6Cl6) were also found to be largely associated with fine adhering soil particles, because of the relatively large surface area per unit mass of these particles. Up to ten-times contaminant enrichment was found when the original soil was compared with the adhering soil.  Soil particles >50 $\mu\text{m}$ were excluded when the soil was dry, implying that the particle size fraction of 0-50 $\mu\text{m}$ is most likely to be ingested involuntarily by children (particles <50-100 $\mu\text{m}$ did not adhere well).  Used to support 250 $\mu\text{m}$ recommendation in 2000 TRW Soil Sampling Guidance.	Sheppard & Evenden, 1994
Agricultural and Brownfield soils  (Saskatchewan and Nunavut, Canada, respectively)	25 adults (14M; 11F)	Soil crumbling/ hand wash  Dry sieved	0.01  <4000 (bulk sample)	Measured dermal adhesion of Canadian soils, found increased adherence with decreasing size, reported metal particle size enrichment, especially for arsenic (420%). Particle size was measured with laser particle size analyzer. The silt fraction (2-50 $\mu\text{m}$ ) is the dominant adhered size. The average particle size of adhered soil varied from 34-105 $\mu\text{m}$ , depending on the soil type. The authors define bulk sediment as <4 mm rather than <2 mm early in the paper; however, 4 mm is only mentioned again in the paper in the caption of Figure 3. The Methods section states the bulk soils were sieved through a 2 mm sieve for measurement of TOC, total metals and presumably, for the adherence experiments. The authors define (concentration) enrichment as (adhered-bulk)/bulk, rather than concentration in adhered fraction / concentration in the bulk soil. Authors recommend that soil samples should be sieved through a	Siciliano et al., 2009

Site-Specific Information	Subjects	Sampling Procedure	Particle Size Tested ( $\mu\text{m}$ ) Moisture Content (% Moisture)	Comments	Reference
				45 $\mu\text{m}$ sieve before estimating risk associated with contaminated soils. Statements about TOC should be carefully reviewed.	
Schoolyard soils  (Japan)	Children (n=10; Nursery school ages)	Hand wash analyzed with laser scattering particle size analyzer  dry sieved	<10 10-100 100-500 500-1000 2000	Found particle size distributions (PSDs) of adhered soils were shifted to the finer fractions, relative to the soil PSDs of the bulk soil; found particles adhered to children's hands are mostly less than 100 $\mu\text{m}$ , while the largest particles were in the 200-300 $\mu\text{m}$ range. Total mass of adhered soil varied greatly between children: mean, median, max = 26.2, 15.2, and 162.5 mg/hand. The average modal diameter was 39 $\mu\text{m}$ +/- 26 $\mu\text{m}$ .	Yamamoto et al., 2006

Table 4. Summary of available of particle enrichment studies.

Site-Specific Information	Sample Preparation	Particle Size Tested ( $\mu\text{m}$ )	Enrichment	Comments	Reference
<p>Tawinteung et al. 2005 (Thailand soils, industrial sources – lead recycling/ smelting)</p> <p>Weiss et al. 2006 (US roadside)</p> <p>Yarlagadda et al. 1995 (manufacturing facility in NYC)</p> <p>Momani 2006 (roadside soils in Jordan)</p>	NA	<p>&lt; 63 <math>\mu\text{m}</math></p> <p>850-2000 <math>\mu\text{m}</math></p>	With particle sizes decreases, lead concentration increased	Review and reanalysis of 4 published studies of lead concentrations and soil particle size, suggests a consistent mathematical relationship between particle diameter and lead concentration enrichment.	Abouelnasr, 2009
<p>Soil samples including: igneous, metamorphic and sedimentary rocks (Murica, Spain)</p>	Air dried, sieved; total elemental composition was determined using ICPAES; mineral and chemical composition was determined by XRD and SEM	<p>&lt; 2mm</p> <p>Three fractions: "clay, silt, sand"</p>	Results showed that Pb, Cd, Cu and Zn are associated with Al and/or Fe-containing minerals such as micas, pyroxenes, and amphiboles and exhibit preferential partitioning into fine particle size fractions in the soils studied.	<p>The study evaluated the contribution of parent geological materials (PGM) to the geochemical composition of the soil, and attempted to establish the distribution and enrichment of major and trace elements (including Pb) in sand, silt and clay particle size fractions.</p> <p>Specific nominal particle sizes were not provided.</p>	Acosta et al., 2011

Site-Specific Information	Sample Preparation	Particle Size Tested ( $\mu\text{m}$ )	Enrichment	Comments	Reference
Urban soils taken from parks (Murcia, Spain)	Surface soils (0-5 cm); bulk samples were air-dried and sieved	850 425 180 150 106 75	Mean Pb content (19 mg Pb/kg) for all bulk samples was 3 times higher than background samples; however, enrichment was not observed with smaller particle size fractions. Consistently high enrichment factors for Pb in the 0-75 $\mu\text{m}$ in two parks were attributed to vehicular depositions.	Metal contents in bulk soil samples are commonly used to assess contamination but metal accumulations in soils are known to increase with decreasing particle size.  We investigated the distribution of metals in various particle size fractions, particle morphology and metal enrichment factors to characterize the properties of soils in four urban parks in Murcia City (SE, Spain).  Soil samples were fractionated and concentrations of Al, Si, Ti, Na, Mg, Ca, P, K, Fe, Cu, Cr, Zn, Pb, Co, Mn and Ni were determined in nine particle size fractions. The contents of metals in playgrounds were below some European cities and low enrichment factors (EF) indicate limited anthropogenic metal depositions. However, The presence of high amounts (60-80 %) of calcite and dolomite in fine fractions might have contributed to metal accumulation through the formation of metal-carbonate complexes. Goethite, especially in PM <sub>10</sub> and PM <sub>2.5</sub> can also serve as sink for metals. The study authors suggested that risk assessment of urban soils in Murcia City (and elsewhere) should be based on (fine) particle size because of the tendency of metal to accumulate in fine particles. In addition, the mineral composition of fine particles should also be considered in risk assessment.	Acosta et al., 2009
Urban soils (Aveiro, Glasgow, Ljubljana, Sevilla and Torino)	Whole Soil	<2 2-10 10-22 22-50 50-2000	Found consistent enrichment in smaller particle size fraction - more than 50% of the total Cu, Pb and Zn accumulate in the <10 $\mu\text{m}$ fraction.  Accumulation in the finer fractions is higher where the overall contamination is lower.	Soils in Torino and Sevilla had increased Pb levels in the 50-2000 $\mu\text{m}$ fractions. Discussed industrialization parameters of the soil sources and speculated that anthropogenic sources of lead contribute to particle size partitioning as demonstrated in soils from the most urban settings.	Ajmone-Marsan et al., 2008
Atmospherically-deposited dust from wide-ranging environments (Riyadh, Saudi Arabia)	Samples collected by ground sweeping, dried overnight and then sieved	20 60 110 170 250 400 750 1500	Metal concentrations increased with decreasing particle size. Using concentration expressed as $\mu\text{g/g}$ of each particle size, Pb in the 20 $\mu\text{m}$ size fraction was enriched 1.5- to 3.0-times compared with the 250 $\mu\text{m}$ size, and 2.5- to 6.4-times compared with the 1500 $\mu\text{m}$ size fraction.	This study evaluated alternative methods for expressing the concentration of metals in particle sizes, using two sets of samples (labeled Group I [n=110] and Group II [n=121]) that were expected to be highly correlated because they were collected, tested, and analyzed under identical conditions. Using the alternative methods for expressing concentration and comparing the particle size concentrations in the two sets of samples, the authors concluded that 1) average concentration of metal in 1 g of a certain particle size gave better correlation than average concentration of metal of a certain size in 1 g bulk soil; and 2) to find correlation between groups fractionated by size, it is important to use the average of particle ratios (calculated as the ratio of the metal concentration per gram of particle of a certain size to the sum of that metal concentration across all particle sizes).	Al-Rajhi et al., 1996a



Site-Specific Information	Sample Preparation	Particle Size Tested ( $\mu\text{m}$ )	Enrichment	Comments	Reference
Outdoor dust samples from pavements or similar surfaces in 231 areas including urban, suburban, rural, motorway, and two industrial sites; indoor dust from 20 public community centers  (Riyadh, Saudi Arabia)	Oven dried	Indoor and outdoor: 20-40 40-80 80-140 Outdoor only: 140-200 200-300 300-500 500-1000 1000-2000	Indoor dust did not show clear pattern of enrichment of Pb concentration at smaller particle sizes.  Outdoor dust showed decrease in concentration with increasing particle size up to 400 $\mu\text{m}$ ; no further decreases were seen at larger sizes.	No additional details on particle size enrichment were provided.	Al-Rajhi et al., 1996b
Residential yard soil samples near an abandoned mine were paired with indoor dust samples; 10 households  (Tucson, AZ, US)	Composite yard samples (surface soil); settled floor dust was vacuumed until achieving 2 g; Oven dried	<63 63-150	Indoor dust was did not show a clear enrichment of Pb concentration at <63 $\mu\text{m}$ ; however, house dust had a much greater GSD. Authors speculated that this is due to house-to-house characteristics.  Outdoor soil had an increased Pb concentration at <63 $\mu\text{m}$ .	All samples were sieved and analyzed for 30 elements via ICP-MS following nitric acid digestion. In house dust, significant differences in concentration were observed for Be, Al, and Mo between particles sizes, with a higher concentration observed in the smaller particle sizes. Approximately 42 and 66% (geometric mean) of soil and house dust mass, respectively, were smaller than 63 $\mu\text{m}$ . Significant differences were also determined for Mg, Ca, Cr, Co, Cu, Ge, Zr, Ag, Ba, concentration in yard soil samples, with the higher concentration observed in the smaller particles size for each element. Authors mention that a questionnaire was administered for each house, but the results were not reported.	Beamer et al., 2012
Street dust collected from pavement, gutter, and road across a roadway	No information in secondary source.	<38 63-125 500-1000	Pb concentration in smallest size fraction (<38 $\mu\text{m}$ ) was 1.4- to 4.8 times higher than corresponding 63-125 $\mu\text{m}$ size fraction (in pavement and gutter samples only; in road sample, enrichment was 0.5) and 6.0- to 13.3 times higher than concentration in corresponding 500-1000 $\mu\text{m}$ size fraction.	As cited in literature review (Fergusson and Kim 1991)	Beckwith et al., 1985
Mining and smelting sites, quarry – river banks and slag  (Coeur d' Alene River Basin, ID)	Adult volunteers (3M, 3F)	Hand washing method with soil  Dry and wet conditions	<63 63-150 150-250 250-2000  Wet: 3.5-14.7%; Dry: <0.25%	Wet media always adhered to a greater extent than dry media. Adhered media generally had higher elemental concentrations than bulk media. Over 60% of the adhered fraction was <63 $\mu\text{m}$ ; this is particularly interesting for the Black Sand Beach because the data show approximately 97% of the BSB material is in the 250 $\mu\text{m}$ - 2mm fraction. While most metals concentrations increased with decreasing particle size, this paper includes some data that show the highest concentration in the bulk soil (e.g., slag material), and others that show little trend with particle size. Regression analyses suggest smaller particle fractions may have higher elemental concentrations. Results of application of a maximum likelihood estimation technique generally indicate that handling of dry media leads to preferential adherence of smaller particle sizes, while handling of wet media does not. Because adhered material can differ greatly in particle size distribution from that found in bulk material, use of bulk concentrations in exposure calculations may lead to poor estimation of actual exposures.	Bergstrom et al., 2011

Site-Specific Information	Sample Preparation	Particle Size Tested ( $\mu\text{m}$ )	Enrichment	Comments	Reference
Total suspended particulate from urban areas (Sicily, Italy)	24-hour air samples collected at low flow (15-20 L/m)	Particles collected on 0.4 $\mu\text{m}$ pore size filters	NA	No information on enrichment by particle size. Reports a range of enrichment values for Pb, S, and Br of 10-1000 relative to crustal composition using Ti as reference element.	Boni et al., 1988
Mine tailings  Review of soil particle size enrichment	NA	40-100 100-200 >200	Concludes that human health risk assessors should look at fractions smaller than 250 $\mu\text{m}$ .	Discusses dermal loading. Discusses enrichment of indoor dust as compared to soil. Discusses differences between sources of contamination and the role of organic matter in bioavailability.	Bright et al., 2006
Residential soils  (Anaconda, Montana)	Children (n=62)  Soil, food and fecal samples	$\leq 250$ <2000	Lead concentration increased with decreasing particle size.	Authors paired particle size data with soil ingestion estimates by using food and fecal marker data. Lead concentrations were estimated with ICP-MS.	Calabrese et al., 1996
Dust, sand, earth collected from roadside gutters and pavements, gardens, schoolyards, and playgrounds  (Manchester, England and Christchurch, New Zealand)	Samples were extracted at various pH	NA	NA	Experiments were designed to evaluate effect of pH on lead extractability from soils. Extractability increased from <10% at pH ~5 to >90% at pH ~1.	Day et al., 1979
Urban street dusts  (London, UK; Manhattan, US; Halifax, Canada; Kingston, Jamaica; Christchurch, New Zealand)	Footpaths, gutters  Dry sieved	<33 33-54 54-88 88-111 111-148 148-165 165-192 192-213 213-963	Increased Pb concentration with decreasing particle size	The metals (Cd, Pb, Cu, Zn, Mn, and Fe) were sequentially extracted from the dust. The overall mass of the dust sampled fell below 200 $\mu\text{m}$ , and <111 $\mu\text{m}$ contained 10-20% of the material for 8 of the samples. Authors state that ~50% of the dust that occurs in the largest particles (213-963 $\mu\text{m}$ ), has a major soil component. Organic content was measured.	Fergusson & Ryan, 1984

Site-Specific Information	Sample Preparation	Particle Size Tested ( $\mu\text{m}$ )	Enrichment	Comments	Reference
		<963			
Street dusts	NA	NA	NA	Review of sources of trace elements in dusts. Reports log enrichment factor (street dust:soil, calculated using cerium as reference element) of ~2 (shown graphically) using median soil concentration data reported in the literature. Reports log enrichment factor for house dust compared with soil of ~1.5.	Fergusson & Kim, 1991
Residential and street dust; garden soil samples  (Christchurch, New Zealand)	3-4 sample/house/10-12 houses/6 areas  Vacuum  Dry sieved	NA	NA	Discusses lead concentration and incidental ingestion of lead. The authors discuss the influence of organic matter, and historical sources of lead. No information on particle size.	Fergusson & Schroeder, 1985
Residential house and street dust; garden soil  (Christchurch, New Zealand)	1 sample/house; 12 houses; 6 areas  Dry sieved	NA	Reports > 3 times enrichment of Pb in house dust and street dust compared with local soils, and > 3 times enrichment in house dust, street dust, and soil compared with crustal abundance.	No information on particle size.	Fergusson et al., 1986
Particles generated by combustion of coal or coal with dried municipal sewage sludge (MSS)  (Stuttgart, Germany)	Particles collected at surface of bag house filter with nominal mean diameter of 3.5 $\mu\text{m}$	Mean diameter of ~3.5 $\mu\text{m}$	Pb concentration was reportedly higher in MSS/coal ash particles than in coal ash alone.	Details of analysis and particle sizes were not reported. Particle size distribution of MSS/coal ash did not differ from coal ash.	Fernandez et al., 2002
Arable soils (tropical)  (Hainan Island, China)	Three composite samples were air-dried and sieved	<53 53-250 250-1000 1000-2000 2000-4000 >4000	The distribution of heavy metals increased with decrease of particle size. The smallest fractions (<53 $\mu\text{m}$ ) occupied only 5.08–9.57%, but had the highest distribution factor for Pb (2.11).	The residues of Pb were correlated positively with the contents of organic carbon as well as Fe in fractions, while a large variation distribution of As was found in particles, indicating its high activity in soil microenvironment.	Gong et al., 2013

Site-Specific Information	Sample Preparation	Particle Size Tested ( $\mu\text{m}$ )	Enrichment	Comments	Reference
<p>Mining (Broken Hill, Port Pirie, Australia; Hobart, Tasmania)</p> <p>Urban Soil (Sydney, Adelaide, Australia)</p>	<p>Soil: top &lt;25 mm - Roadside</p> <p>Dusts: vacuum cleaner dust; surface wipe; dust fall</p>	<p>&lt;5 5-10 10-38 38-53 53-75 75-150 150-250 250-500 500-1000 &gt;1000</p>	<p>Considerable variation was observed in the weight percentages for the various fractions</p> <p>Finer fractions contain 2-9 times higher concentration than bulk fractions</p>	<p>Authors suggest that it is necessary to analyze the bulk fractions as well as the &lt;150 <math>\mu\text{m}</math> (or even better, the &lt;100 <math>\mu\text{m}</math>) fraction for soils and the &lt;100 <math>\mu\text{m}</math> fraction for dusts.</p>	<p>Gulson et al., 1995</p>
<p>Small arms shooting ranges, incinerators, historical fill material, mining and smelting, and gasworks  (Australia)</p>	<p>16 soil samples</p>	<p>&lt;50 &lt;100 &lt;250 &lt;2000</p>	<p>Increasing concentration with decreasing particle size.</p> <p>&lt;50 <math>\mu\text{m}</math> fraction, the average concentration of lead was approximately twice the average concentration of lead in the &lt;2 mm fraction but varied between 0.86 - 5.31.</p> <p>Lead enrichment in the &lt;50 <math>\mu\text{m}</math> particle size fraction was up to 5 times the concentration observed in the bulk soil.</p>	<p>The slopes and coefficient of determinations (<math>R^2</math>) reported by the authors for regression models of lead concentration in particle size intervals (i.e., &lt;50, &lt;100 and &lt;250 <math>\mu\text{m}</math>) on lead concentration in the bulk sample (i.e., &lt;2 mm fraction) appear to be highly influenced by the two samples collected from a mining/smelting site.</p> <p>This paper also reports data on bioaccessibility, determined using the Solubility Bioavailability Research Consortium (SBRC) in vitro assay (3 replicates for the three grain size fractions provided above for each of the 16 soils). They report trends of increasing bioaccessibility with decreasing particle size fraction for 9 of the 16 soils; 6 of the 9 were statistically-significant trends. The authors compare IEUBK-predicted PbB levels to measured PbB levels for two of the mining/smelting sites. The comparisons are made at three particle size fractions, with and without adjusting the IEUBK model bioavailability parameter.</p>	<p>Juhasz et al., 2011</p>
<p>Mine tailings, unprocessed waste rock, background samples  (Randsburg, CA, US)</p>	<p>40 soil samples</p>	<p>&lt;20 20-32 32-45 45-75 75-125</p>	<p>Increased Pb concentration with decreased particle size</p> <p>&lt;250 <math>\mu\text{m}</math> collectively comprise 25.3-61.4% of the samples' mass;</p>	<p>Authors suggest incidental ingestion of adhered particles in the &lt;250 <math>\mu\text{m}</math> fraction</p>	<p>Kim et al., 2011</p>

Site-Specific Information	Sample Preparation	Particle Size Tested ( $\mu\text{m}$ )	Enrichment	Comments	Reference
		125-250 250-500 500-1000 1000-1700 1700-2830 >2830			
Chromium-contaminated site  (New Jersey, US)	21 soil samples  Dry sieved; XRF analysis	<400 250-400 175-250 75-175 45-75 38-45 <38 10 2.5 1.0	Increased Pb levels with decreased particle size (<38 $\mu\text{m}$ ).	The study focuses on the inspirable, thoracic and respirable particles that would enter the respiratory tract. Particles were resuspended to determine size fractions <38 $\mu\text{m}$ . ICP-AES was used to extract chromium. Enrichment factors for lead and chromium were determined by XRF analysis.	Kitsa et al., 1992
Urban playgrounds  (Uppsala, Sweden)	Wet sieved	<50 50-100 <4000	Demonstrated average 1.5 times enrichment for metals in finest fraction (<50 $\mu\text{m}$ ) compared with either the bulk sample or the 50-100 $\mu\text{m}$ fraction.	For two soils with high (58%) and low (13%) sand content, Pb was enriched in smaller size particles (1.6- to 2.7 times) in high sand soils but was not enriched by size in low sand soils. Metals content inversely correlated with sand content.	Ljung et al., 2006
Urban playgrounds  (Uppsala, Sweden)	Assessed for in vitro bioaccessibility using a three compartment digestion model.	<50 <4000	Lead levels were consistently higher in <50 $\mu\text{m}$ fraction	It was concluded that in soils with low metal load and sufficient number of binding sites, the effect of particle size range is diminished.  "The difference in bioaccessibility between elements was concluded to be due to differences in origin, sorption behavior and pH dependence. The study also found that the bioaccessible amount of metal in ingested soil is not always related to particle size or to soil mass in soils with low contaminant levels. Factors such as pH dependence of the metal and the soil's clay content are also significant in determining bioaccessibility."  The particle size range only had an effect on Pb bioaccessibility when there was limited access to binding sites. With a clay content >10%, the strong pH dependence of Pb was found to have a greater effect on the distribution of bioaccessible Pb. As suggested before, the preference for sorption to clay particles diminishes the effect of particle size range in unpolluted soils, since most metal ions are bound to these small particles, which were present in both scenarios.	Ljung et al., 2007

Site-Specific Information	Sample Preparation	Particle Size Tested ( $\mu\text{m}$ )	Enrichment	Comments	Reference
				However, when the number of clay particles was limited, an effect of particle size was observed.	
Urban soils  (Hong Kong)	8 soil samples  Wet sieved	<2 2-10 10-50 50-100 100-280 280-2000	2-10 $\mu\text{m}$ had increased metal concentrations; 50-280 $\mu\text{m}$ less marked	Variability in distribution of contaminants by particle size fraction supports argument against generalization.  Reported increased bioaccessibility with finer particle size, - there was no measure of bioavailability in this study. "The mobility, bioavailability, and human bioaccessibility of Pb and Zn in bulk soils correlated significantly with metal concentrations in fine silt and/or very fine sand fractions."	Luo et al., 2011
Urban soils  (Torino, Italy; Sevilla, Spain)	10 soil samples  Dry sieved  Bioaccessibility (SBET, EDTA)	<2 2-10 10-22 22-50 50-2000	Particle sizes <50 $\mu\text{m}$ had generally higher lead concentrations than >50 $\mu\text{m}$	In soil samples from both cities, the finest fractions had the highest EDTA extractability for lead and lowest SBET lead bioaccessibility as compared to other fractions and whole soil. The authors suggest the differences in bioaccessibility between the fractions are due to the presence of humic content.	Madrid et al., 2008
Urban soils/street dust  (Al Zarqa, Jordan)	18 soil samples	<63 63-125 125-250 250-600	<63 $\mu\text{m}$ had the greatest Pb concentrations	Samples were obtained from the sides of busy roads where leaded gas still in use	Momani, 2006
Mining  (Coeur d' Alene River Basin, ID, US)		<175		The rationale for this particular sieve size includes compatibility with earlier soil sampling protocols in the Coeur d'Alene River basin and consistency with soil adherence data for dermal exposures.  Although enhanced lead enrichment would be expected in soils processed with the 175 $\mu\text{m}$ sieve instead of the 250 $\mu\text{m}$ sieve, the real issue from a human exposure assessment standpoint is not lead enrichment, but rather the accurate characterization of lead in the particles that play the dominant role in the soil/dust-to-hand-to-mouth pathway.  Data derived for analysis of Bunker Hill exhibited acceptable agreement between observed and predicted PbB levels for 15 consecutive years using model inputs based on soil and dust samples sieved to less than 175 $\mu\text{m}$	NRC, 2005
Sediment samples near the River Avon	17 soil samples	<20 20-63	Mean and maximum Pb concentrations increased with decreasing particle size fraction.	Authors state, "It was concluded that, while in certain specific circumstances it may be most appropriate to make forensic comparisons based on a very narrowly defined particle size range, for the majority of purposes the <150 $\mu\text{m}$ fraction	Pye et al., 2007

Site-Specific Information	Sample Preparation	Particle Size Tested ( $\mu\text{m}$ )	Enrichment	Comments	Reference
(United Kingdom)	Wet and dry sieved	63-150 150-2000		provides the best compromise between sample size requirements and data resolution."  Also includes mean, max, min and variance (coefficient of variation) for 49 metals and oxides, and multivariate statistical methods that could be useful.	
Agricultural and Brownfield soils  (Saskatchewan and Nunavut, Canada, respectively)	13 agricultural soil samples  17 Brownfield soil samples  Dry sieved	0.01 <2000  <4000 (bulk sample)	Increasing metal concentrations with decreasing particle size fractions.	Measured dermal adhesion of Canadian soils, found increased adherence with decreasing size, reported metal particle size enrichment, especially for arsenic (420%). Mean dermal adherence for agricultural and Brownfield soils were 34 $\mu\text{m}$ and 105 $\mu\text{m}$ , respectively. Particle size was measured with laser particle size analyzer. The silt fraction (2-50 $\mu\text{m}$ ) is the dominant adhered size. The average particle size of adhered soil varied from 34-105 $\mu\text{m}$ , depending on the soil type. The authors define bulk sediment as <4 mm rather than <2 mm early in the paper; however, 4 mm is only mentioned again in the paper in the caption of Figure 3. The Methods section states the bulk soils were sieved through a 2 mm sieve for measurement of TOC, total metals and presumably, for the adherence experiments. The authors define (concentration) enrichment as (adhered-bulk)/bulk, rather than concentration in adhered fraction / concentration in the bulk soil. Authors recommend that soil samples should be sieved through a 45 $\mu\text{m}$ sieve before estimating risk associated with contaminated soils. Statements about TOC should be carefully reviewed.	Siciliano et al., 2009
Urban soils (shooting range, incinerator, historical fill, mining smelting, gaswork activities)  (Australia, New Zealand)	31 soil samples  Dry sieved	<250  <2000	Pb concentration in the <250 $\mu\text{m}$ fraction was greater than the <2mm fraction.	Demonstrated variable enrichment between the size fractions. Used in vitro assay to measure lead bioaccessibility in the <250 $\mu\text{m}$ (dry sieved) fraction only. Compared the bioaccessibility using either USEPA Pb IVBA method or SBRC assay. Demonstrated wide variety of gastric phase lead bioaccessibility 35-105%.	Smith et al., 2011
Battery recycling plant  Lead-smelting factory  (Thailand - sandy loam, sandy clay, clay)	6 soil samples  Surface soil (0-15 cm)  Subsoil (15-30 cm)	<150 150-250 250-2000	Pb concentration was ~2 times higher in the <150 $\mu\text{m}$ than in the 250 $\mu\text{m}$ -2mm fraction.	The primary objective of this study was to characterize the chemical forms of lead in soil that affect the extraction of lead from the soil; however, the paper includes data on enrichment of lead from 6 samples collected from three locations (two contaminated areas and one reference area) and two depths below the ground surface.	Tawinteung et al., 2005
Urban roadway "grit"  (8 sites, New York City, NY, US)	225 soil samples  Dry sieved	<63 63-125 125-250 250-500 500-2000	84% of the particles were in the range of 125-500 $\mu\text{m}$ . The highest concentration of lead was in the smallest fraction analyzed (<63 $\mu\text{m}$ ).	Pb concentrations ranged from 20-7460 ug/g.	Weiss et al., 2006

Site-Specific Information	Sample Preparation	Particle Size Tested ( $\mu\text{m}$ )	Enrichment	Comments	Reference
		2000-4000			
Bulk surface soil samples obtained near five industrial facilities and along roadsides  (5 locations in California, US)	Soil was collected up- and down-wind of each facility; samples of the soil were resuspended in a laboratory chamber to generate $\text{PM}_{10}$	$\text{PM}_{10}$  <38 (fine soil)  300-2000 (coarse soil)  Bulk soil	Pb in $\text{PM}_{10}$ was enriched by 1.10 to 8.31 times compared with bulk soil samples  Pb in fine soil was enriched by 1.12 to 7.83 times compared with coarse soil samples	Pb in $\text{PM}_{10}$ was enriched 5.36 – 88.7 times compared with uncontaminated California soils (highest enrichment value was associated with a Pb smelter site).  The geometric mean particle diameters of the bulk soil samples ranged from 41.2 to 344 $\mu\text{m}$ .  Facilities included the following: glass manufacturing, perlite mining, borax processing, lead smelter, sandblasting, and roadside.	Young et al., 2002



## APPENDIX A. Sample Preparation and Analytical Considerations

The TRW Lead Committee (TRW) recommends the following procedures for sample preparation and analysis of lead in soil and dust at Superfund sites to improve the representativeness of samples used to estimate concentrations of ingested lead in soil and dust. There may be site-specific instances where the recommendations below do not apply, such as sites where the soil is typically wet (larger particles may adhere to skin when the soil is wet). Further information on soil sampling is available from U.S. EPA (2013, 2003).

After drying, soil or dust samples are passed through a No. 4 (4.75 mm) or a No. 10 (2.0 mm) sieve (ASTM, 1999) to remove any large debris. The resulting material is referred to as the total sample. The total sample is weighed and sieved through a No. 100 W.S. Tyler® sieve to identify the “coarse” (>150 µm) and the “fine” (<150 µm) fractions. The fine fraction may be further fractionated if site-specific circumstances warrant.

If only one analysis is to be performed on a soil or dust sample at a lead-contaminated site, as is sometimes the case at a removal site, the TRW Lead Committee recommends analyzing the lead concentration in the fine fraction (fraction which passes through a No. 100 W.S. Tyler® mesh sieve) at a minimum, with site-specific consideration for the need for further assessment. The particle size fraction used should provide the most accurate characterization of the current risk from exposure by incidental ingestion at the site.

To account for the potential for enrichment of lead, the concentration of lead should be analyzed in both the fine and coarse fractions, at least for a subset of samples. After determining the concentration of lead in the coarse and fine particle size fractions, the lead concentration of the total sample may be reconstructed using a weighted average of the coarse and fine fractions. The resulting ratio (*i.e.*, the enrichment ratio) between the concentrations of lead in the fine fraction relative to the concentration in the total sample may be used to develop a site-specific “adjusted” cleanup level that would be applicable to total soil sampling data if the data supports an assumption that the enrichment ratio is constant across the site or within an exposure unit. In addition, if prior soil sampling data are available, such analysis may allow for comparison with earlier sampling data.

When there is potential for the total sample to contain higher concentrations of lead than the fine fraction (*e.g.*, if coarse material from mining or industrial operations contains higher concentrations of lead than the fine fraction), the future degradation of these coarser materials into finer particles should also be considered (*e.g.*, addressed by using the total soil concentration for the risk assessment of potential future exposures). In addition, total sample concentrations represent deliberate soil or dust ingestion (Lenoir et al., 1997). In these instances, at least 20% of the surface soil samples, or a minimum of 20 samples, should be analyzed for lead concentration in both the coarse (>150 µm) and the fine (<150 µm) particle size fractions. This recommendation to consider the lead concentration in larger particle size fractions may be particularly relevant to sites where large particles of lead may be present in soil, such as shooting ranges or battery recycling operations.

While the sieving of all samples is preferred, at some sites this may not be practical. When sieving is limited, a constant (*e.g.*, relative standard deviation [RSD] <30%) enrichment ratio across sampling locations, the concentration of lead in the fine fraction may be used to estimate the concentration of lead in the total sample. For this estimation, the TRW Lead Committee recommends using a statistical regression model (*i.e.*, full regression analysis) to examine the relationship between concentrations of lead in the different soil fractions.

The TRW Lead Committee recommends assistance from a statistician to develop and evaluate such regression models<sup>8</sup>. Unless prediction errors are relatively small (*e.g.*, RSD <30%) (10–20% of the best estimates), the TRW Lead Committee further recommends that upper prediction limit to estimate concentrations of lead in the fine fraction be used for site applications. Large prediction errors indicate that the concentration in the fine fraction should be measured rather than predicted using a regression model, particularly if the predicted concentration of lead in the fine fraction is close to the risk management decision range.

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<sup>8</sup> Regression models often provide the best estimates of lead concentrations in the fine fraction (*i.e.*, the regression line) and predict errors about the regression line. In some instances, however, mixed models may be needed.