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Particle size distributions, size concentration relationships, and adherence to hands of selected geologic media derived from mining, smelting, and quarrying activities ☆

Carolyn Bergstrom, Jeffry Shirai, John Kissel *

University of Washington, Department of Environmental and Occupational Health Sciences, United States

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ABSTRACT

Hand-to-mouth activity, especially in children, is a potentially significant pathway of exposure to soil contaminants. Hand-mouthing behavior is of particular concern in areas impacted by mining, smelting, and quarrying activities as these activities may lead to elevated levels of heavy metals in soil. In order to estimate potential exposures to contaminated geologic media attributable to hand-to-mouth contact, it is useful to characterize adherence of those media to skin, as contaminant concentrations in adhered media may differ greatly from unfractionated, whole media concentrations. Such an investigation has been undertaken to aid estimation of exposures to arsenic, cadmium, lead, and zinc in nine different geologic media collected in the Pacific Northwest region of the United States. After establishing the particle size distribution of each medium (fractions <63 μm, 63–150 μm, 150–250 μm, and 250 μm–2 mm were determined) and target elemental concentrations within each particle size fraction, an active handling protocol involving six volunteers was conducted. Wet media always adhered to a greater extent than dry media and adhered media generally had higher elemental concentrations than bulk media. 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. Since lead has historically been a metal of particular concern, EPA's Integrated Exposure Uptake Biokinetic (IEUBK) Model was used to examine the potential consequences of evaluating ingestion of the selected media assuming concentrations in adhering versus bulk media.

1. Introduction

Children often come into contact with soil during recreational activities and may be exposed to agents in the soil via both ingestion and dermal absorption. Increased body burden following exposure to environmental contamination has been observed in multiple studies. For example, positive correlations between biomarker levels in young children and either proximity to a source or concentration in soil or dust have been reported for both lead (Lanphear et al., 1998; Schmitt et al., 1979) and arsenic (Hwang et al., 1997; Polissar et al., 1990). In the case of inorganic elements or compounds, for which dermal absorption is generally assumed to be slow, soil ingestion is the more likely explanation for elevated exposures. The media examined here are derived from mining, smelting and quarrying activities and contain high concentrations of inorganic elements. Therefore this investigation is limited to characterization of properties of media adhering to hands.

1.1. Elemental and soil adherence data

Early efforts to characterize soil loads on hands have been summarized elsewhere (Kissel et al., 1996a). In several of those studies direct measurement was of mass of lead rather than soil. Notably Roels et al. (1980) investigated dermal adherence of lead in the environment by comparing blood lead levels of children attending schools near and far from a smelter in rural and urban environments of Belgium. They sampled air and soil lead concentrations in both areas. Their findings suggest that lead ingestion from hand contamination contributes greatly to blood lead levels when the amount of lead on hands is higher than 20 μg/hand. Specifically, they suggest that lead intake from contaminated hands contributes two to four times more to blood lead levels than lead inhaled through the air.

Subsequently a series of field (Holmes et al., 1999; Kissel et al., 1996a; Shoaf et al., 2005a,b), laboratory (Holmes et al., 1996; Kissel et al., 1996b), and greenhouse (Kissel et al., 1998) studies were conducted

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* Corresponding author at: 4225 Roosevelt Way NE Ste. 100, Box 354695, Seattle, WA 98105, United States. Tel.: +1 206 543 5111; fax: +1 206 543 8123.
E-mail address: jkissel@uw.edu (J. Kissel).

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to characterize soil adherence on multiple body parts in both children and adults. Field trials have revealed higher loads on palms and bare feet than on other body parts. In addition, the duration of activity does not appear to accurately predict soil loading (Kissel et al., 1998; Shoaf et al., 2005b). This may be because initial contact results in high amounts of surface loading, but during subsequent activity, abrasion and movement may actually remove soil from the skin. Generally, wet soil adheres to a greater extent than dry soil under both laboratory and field conditions. Preferential adherence of finer particles is observed under dry loading conditions (Kissel et al., 1996b). USEPA summarized much of the soil adherence literature in its guidance material for dermal risk assessment USEPA (2007). Two papers that were not included in that review are also relevant here.

Choate et al. (2006) investigated dermal adherence of a clay loam collected from Fort Collins, Colorado as well as a silty-clay loam collected from Ames, Iowa. Adult volunteers' hands were brought into contact with bulk soil in a passive handling protocol. Choate et al. observed that particles of dry or moderately moist soils adhering to hands were generally less than 63 μm in diameter. They also studied the effect of moisture content on soil adherence and observed that higher media loading occurred following contact with soil of higher moisture content.

In a laboratory experiment in which an adult subject pressed a hand into soil, Yamamoto et al. (2006) observed that finer soil particles adhered more efficiently to the hands. In addition, in a trial wherein wet hands came into contact with the soil, there was an increase in the amount of adhered soil. Their results also suggest that contact between the driest hands and soil resulted in the smallest particle sizes adhering. Yamamoto et al. also analyzed soil adhering to children's hands following activities at a playground and schoolyard outside a nursery in Kanagawa Prefecture in Japan. They compared the particle sizes adhering to hands to the size distribution of the unfractionated soil and found that finer particles adhered to hands. They observed that soil particles larger than a few hundred microns did not adhere to the children's hands.

1.2. Elemental concentration enrichment

Davies and White (1981) studied soil pollution in the vicinity of a lead mine in the U.K.; a clear tendency for increased lead concentrations in smaller particles of mine waste was observed. Fractions investigated were: <64 μm, 64–140 μm, 140–200 μm, 200–1000 μm, and 1000–2000 μm. Average lead concentrations of the aforementioned particle sizes were 76.1, 36.8, 24.2, 14.1, and 16.4 μg/g, respectively. These are the means of three samples, except for the 1000–2000 μm fraction which is the mean of two samples. The smallest particle size fraction displays a lead concentration almost five times higher than the largest particle size fraction.

Sheppard and Evenden (1992) investigated impacts of erosion on particle size distribution and contaminant concentrations in a silty clay loam and a loam collected near Pinawa, Manitoba. Fractions greater than 200 μm in diameter were found to have two- to eight-fold *lower* concentrations of uranium, thorium, and lead than the fractions with diameters less than 2 μm. Sheppard and Evenden suggest that use of bulk soil concentrations for adhering soil in inadvertent soil ingestion scenarios may underestimate impacts by approximately 30%.

Sheppard and Evenden (1994) also observed that particles with smaller diameters adhere to skin and that adhering soil is more contaminated than bulk soil. The size of the soil fractions ingested may determine the mass of elements ingested. This is due in part to a larger reactive surface area per unit mass in particles with smaller diameters, leading to increased elemental concentrations (Sheppard, 1995).

Gulson et al. (1995) analyzed lead concentrations in soil and dust in the vicinity of an Australian lead smelter in soil. Finer particle sizes had 2–9 times higher concentrations than bulk fractions. However, only two particle sizes were analyzed: <2 mm and 38–53 μm. In the analysis of house dust, more particle size fractions were analyzed: 53–75 μm, 75–150 μm, 150–250 μm, 250–500 μm, and <2 mm. Lead concentrations in the smaller particle size fractions were up to three times that found in the bulk dust sample.

Sheets and Bergquist (1999) investigated soil at a Superfund site contaminated from salvage activities and metal recycling. They found generally increasing concentrations of lead in the smallest particle size fractions under study (fractions included > 4.75 mm, 4.75–2.0 mm, 2.0–0.425 mm, 0.425–0.074 mm, and < 0.074 mm).

Ljung et al. (2006) investigated childhood exposures to elements in soil from playgrounds in urban Uppsala, Sweden. The areas were of differing land use including roadsides, former and current industrial land, natural lands, and city centers. The soil was wet-sieved into three fractions: < 4 mm (representing deliberate ingestion), 50–100 μm, and < 50 μm (representing involuntary ingestion). They found that particles in the < 50 μm size fraction had approximately 1.5 times higher As, Cd, Cr, Pb, and Zn concentrations than those in the < 4 mm or the 50–100 μm fractions. Furthermore, they found that 30–35% of the As and total metal contents were in the < 50 μm particle size fraction. Statistical correlation between sand content and elemental concentrations was carried out and it was found that decreased sand content correlated with increased metal and As contents.

1.3. Summary

Soil adherence, particle size and elemental concentration enrichment, hand-to-mouth behavior, and soil ingestion behavior are important considerations when determining children's exposure to contaminated soils. Prior studies have reported both preferential adherence of fines and elemental concentration enrichment in fine fractions. The study reported here is unique in that it involves geologic media that are not traditional soils and that it employs adherence using a dynamic handling protocol. The authors hypothesize that media adhered to hands have larger concentrations of potentially dangerous elements than bulk media, which would be the case if smaller particles have higher elemental concentrations and smaller particle sizes preferentially adhere to hands.

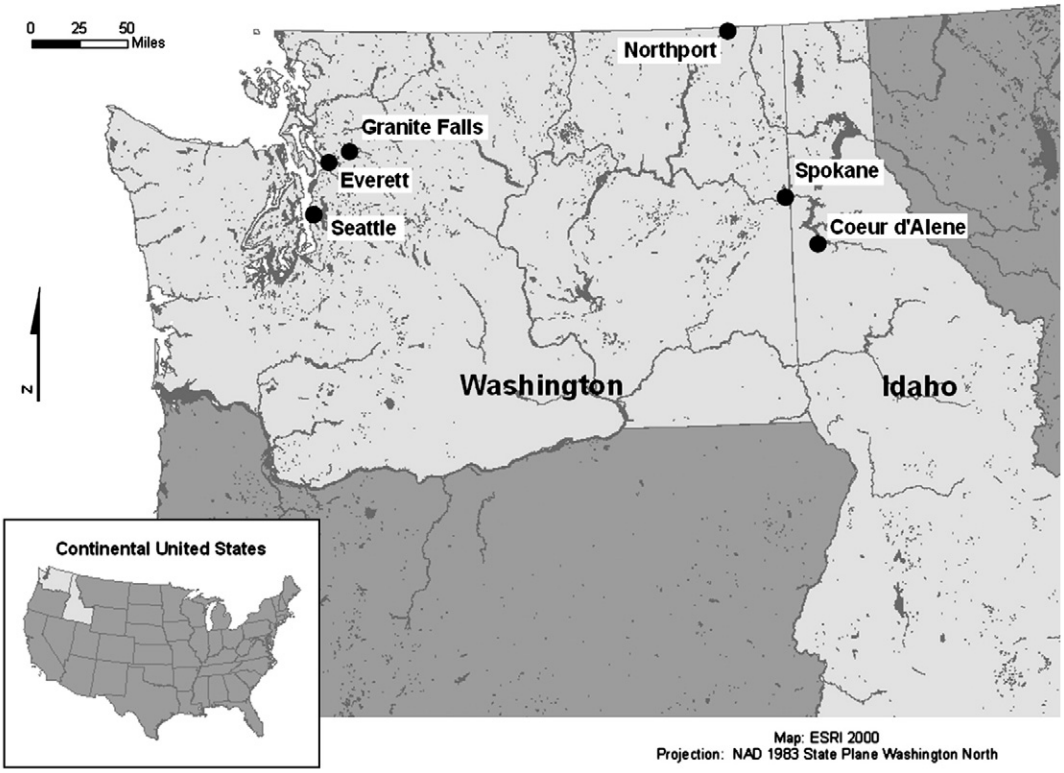
2. Methods

2.1. Study design

Nine media samples were collected from areas impacted by mining and smelting, and from a quarry. All were taken from locations that have attracted some degree of regulatory attention. Reference soil samples were collected from a residential lot assumed to be uninfluenced by the aforementioned activities. All samples analyzed were collected from locations in the Pacific Northwest region of the United States (see Fig. 1) between 2006 and 2008.

2.2. Description of sample locations

Three samples (CDA-1, -2, and -3) were collected from locations (sand bar, alluvial beach, and river bank, respectively) along the Coeur d'Alene River in Idaho. All three samples were obtained downstream from the Bunker Hill Mining and Metallurgical Complex, placed on the EPA's National Priorities List in 1983. A fourth sample analyzed (called Spokane River) was collected from a bank of the Spokane River in Eastern Washington. This river drains Coeur d'Alene Lake and is impacted, although to much less of a degree, by the same mining activity that is associated with samples CDA-1, -2, and -3. A sample called Black Sand Beach (BSB) was acquired 7 miles northeast of Northport, Washington along the Columbia River. BSB was sampled downstream from a lead and zinc smelter located in Trail, British Columbia (Canada). The Everett slag sample, originally placed as roadbed fill, was collected from an exposed roadside near the former Asarco smelter in Everett, WA. IMQ 3/8 minus, IMQ 3/4–3/8, and IMQ



Map: ESRI 2000
Projection: NAD 1983 State Plane Washington North

Fig. 1. Map of approximate geographic locations of sampling areas.

3/4 minus are quarried gravel products obtained from Iron Mountain Quarry, Inc. in Granite Falls, Washington. Finally, Reference soils 1a and 1b were acquired from a backyard landscaping project in Seattle, WA. These yard soil samples served as control samples, as they were not expected to have been impacted by the industrial activities mentioned above.

2.3. Particle size distribution in media

All of the above samples were sieved initially with a 0.75-inch (19 mm) ASTM sieve (coarse series) to remove rocks and debris. Materials retained on this sieve were not used in any subsequent analyses. Two separate aliquots (each approximately 0.35 L) of each media were sieved into five particle fractions with a sieve stack (W.S. Tyler, U.S. standard series; ASTM nos. 10 (2 mm), 60 (250 μm), 100 (150 μm), and 230 (63 μm)). The sieve stack was agitated on a mechanical sieve shaker (ELE International) for 10 min. After sieving, each particle fraction was weighed to determine its proportion of the whole sample.

2.4. Chemical analysis

Aliquots (250 mg) of each size fraction (<63 μm, 63–150 μm, 150–250 μm, 250 μm–2 mm, and >2 mm) of each media were analyzed by the University of Washington's Department of Environmental and Occupational Health Sciences Environmental Health Laboratory by inductively coupled plasma mass spectrometry (ICP-MS) (using a modified version of EPA Method 6020A) for arsenic (As), cadmium (Cd), cobalt (Co), chromium (Cr), lead (Pb), and zinc (Zn) concentrations. Three or four aliquots were analyzed depending on available mass of media in each size fraction. The largest size fraction was pulverized in a ball mill (SPEX SamplePrep 8000, SPEX CertiPrep) prior to microwave assisted acid digestion (using a modified version of EPA Method 3051A). Only As, Cd, Pb, and Zn are reported and discussed here as lab results for Cr and Co were both relatively low and variable. The geometric mean reconstructed bulk concentrations of Cr and Co across all media were 11.6 and 10.8 μg/g, respectively.

2.5. Media adherence to hands

Six adult volunteers (three females and three males) actively handled approximately 1.5 cups of each unfractionated medium placed in a plastic trash bag (Glad ForceFlex®) for 30 s under wet and dry conditions. The pull strings of the trash bag were tied tautly around the volunteer's wrists and they were asked to actively handle the media in a handwashing motion. Volunteer information is reported in Table 1.

Wet conditions were obtained by adding deionized water to the media in the plastic bag immediately before handling and massaging the bag by hand to ensure thorough mixing. Mean moisture contents in the wet trials ranged from 3.5% for IMQ 3/8 minus to 14.7% for the Spokane River media. Variation in moisture content in the wet trials reflects the variable moisture holding capacity of the media examined here. In the dry trials, mean moisture content was below 0.25% for all media. After carrying out the handling protocol, subjects' hands were washed with deionized water and if needed a <2% solution of Liquinox soap (Alconox) over a stainless steel baking pan (Polar Ware). The

Table 1
Summary of pertinent volunteer characteristics.

Sex	Average mass (kg)[a]	Average height (cm)[a]	Average hand surface area (cm^2)[a]	Sample group [b]
Female	61.3	166	821	A
Male	90.1	187	1083	A
Female	66.8	167	850	B
Male	92.5	187	1099	B

[a] Average of three volunteers.

[b] Sample group A includes CDA 1–3, BSB, Spokane River, IMQ 3/8 minus, Everett slag, Reference soil 1a. Sample group B includes IMQ 3/4–3/8, IMQ 3/4 minus, and Reference soil 1b.

wash water was filtered through 47-mm diameter mixed cellulose ester (MCE) membrane filters with nominal pore size of 0.45 µm (Zefon International) and secured over stainless steel manifolds (Nalgene). Ten milliliters of the filtered water for each sample were submitted for ICP-MS analysis to determine if substantial masses of elements bypassed the filter. The media-laden filters were placed in tared aluminum weigh boats (VWR) and oven dried (Labline, Barnstead International) overnight at 100 °C. Filters and weigh boats were removed from the oven, cooled, and weighed on an analytical balance (Mettler College 150). Filtrate was acidified prior to analysis for elements by ICP-MS.

After obtaining elemental concentration data for media-laden filters, elemental loading on volunteers' hands was determined using estimated hand surface area. Surface area was calculated as:

$$SA_{\text{hands}} = a0 \cdot W^{a1} \cdot H^{a2}$$

where a0 = 0.0131 (females) or 0.0257 (males), a1 = 0.412 (females) or 0.573 (males), a2 = 0.0274 (females) or −0.218 (males), W = weight (kg), and H = height (cm). The values used for a0, a1, and a2 are based on gender-specific regression equations generated for adults. The above equation is recommended in U.S. EPA's Exposure Factors Handbook USEPA (1997).

2.6. Statistical analysis

SPSS® 17.0 was used to carry out statistical analyses. Lognormality and normality of media loading for all geologic media sampled were evaluated using the Lilliefors modification of the Kolmogorov–Smirnov test as well as the Shapiro–Wilk test. Regression analyses were used to examine trends between log-transformed particle size and non-transformed elemental concentration.

Proportions of particle size fractions adhering to hands were estimated using a Maximum Likelihood Estimation (MLE) technique. The Solver tool in Microsoft® Excel was used to carry out this task. Solver performs optimization within predefined constraints. Fractions of adhering mass attributed to each size fraction were allowed to vary to minimize a function representing the difference between predicted and observed concentration in adhering media mass. This exercise was conducted for each media sample for both wet and dry trials.

2.7. Integrated Exposure Uptake Biokinetic (IEUBK) Model

The EPA's 2009 IEUBK Model was used to estimate potential incremental changes in blood lead levels in children two to five years old if they consumed soil with lead concentrations found in sample collection areas. The impact of using bulk media lead concentrations versus adhered media lead concentrations was investigated.

3. Results/discussion

3.1. Particle size characteristics of media

Table 2 displays the particle size characteristics of the 11 geologic media investigated by percent of total mass. Several of the samples have a markedly uneven distribution of particles across the size fractions. For example, all three IMQ samples as well as the Everett slag sample have greater than 50% of their mass in the >2 mm particle size fraction. In addition, over 97% of the BSB sample mass fell in the 250 µm–2 mm size class. The IMQ sample size distributions reflect deliberate production for commercial purposes. Both the BSB and Everett slag particle size distributions are at least partially attributable to industrial smelting. The BSB sample's particle size distribution may also reflect hydrodynamic sorting.

Table 2
Particle size distributions for all media by percentage.

	% of each fraction making up whole sample by mass				
	<63 µm	63–150 µm	150–250 µm	250 µm–2 mm	>2 mm
CDA 1	15.1	50.0	27.6	5.46	1.77
CDA 2	8.23	37.8	45.6	8.29	0.0112
CDA 3	29.5	34.0	14.3	10.7	11.5
BSB	0.111	0.580	1.93	97.1	0.276
Spokane River	7.18	15.0	18.3	44.5	15.0
IMQ 3/8 minus	3.10	3.96	3.66	32.3	56.9
IMQ 3/4–3/8	0.369	0.771	0.465	0.483	97.9
IMQ 3/4 minus	0.983	2.52	2.81	6.23	87.4
Everett slag	9.01	7.27	5.99	18.2	59.6
Reference soil 1a	3.54	9.66	25.1	50.4	11.3
Reference soil 1b	3.98	9.19	24.8	56.2	5.83

3.2. Concentrations in different particle fractions

Table 3 presents concentrations of As, Cd, Pb, and Zn in the five particle fractions under study. Concentrations are not reported for the >2 mm fraction of CDA-2 because there was an insufficient mass of particles to permit chemical analysis. The values reported for the >2 mm fractions of CDA-1 and BSB are the geometric means of three aliquots, whereas all remaining fractions are geometric means of four aliquots. This is due to a lack of sieved mass rather than total absence of coarse particles in the medium. Maximum concentrations of Zn (86,300 ppm, >2 mm fraction), As (2040 ppm, <63 µm), and Pb (29,400 ppm, >2 mm) were found in Everett slag. The highest Cd concentration was found in CDA-3 (98.5 ppm, >2 mm fraction).

Forty-four linear regression analyses were carried out (4 elements × 11 media) to determine if there was a particle size concentration effect. Eight datasets had a statistically significant negative correlation (smaller particle sizes had a higher elemental concentration). Twenty-seven of the datasets had a non-statistically significant negative correlation while the remaining nine datasets revealed a non-statistically significant positive correlation. No data set had a statistically significant positive correlation. Statistical significance was defined as a regression slope having 95% confidence bounds that exclude zero.

Considering only As and Pb (which are of primary interest on human health grounds), 7 of the 22 regression analyses resulted in statistically significant negative correlations. Twelve of the 22 resulted in non-statistically significant negative correlations, and 3 in non-statistically significant positive correlations. In summary, 86% of the regression analyses carried out for As and Pb resulted in a negative correlation (either statistically or non-statistically significant).

3.3. Media adherence

Table 4 displays media loading across all 11 unfractionated samples, for dry and wet handling trials. In the Shapiro–Wilk test, normality and lognormality could not be rejected in 43 of 44 data sets (11 media, male and female, wet and dry). Hence, lognormality was assumed for all remaining statistical tests consistent with prior precedent (Holmes et al., 1999, Kissel et al., 1998). Independent samples t-tests were conducted on 22 data sets to determine if there was a statistical difference between media loading among men and women. All 22 tests were nonsignificant ($p > 0.05$). Lognormality within each condition for each media (across genders) was tested using the Lilliefors modification of the Kolmogorov–Smirnov test. Lognormality could not be rejected for 21 out of 22 data sets. Subsequently, male and female data were lumped within each condition (wet or dry) for each sample and paired samples t-tests were carried out. In 11 of 11 media, media mass adhering after wet trials was significantly higher than after dry trials ($p < 0.01$).

The analysis of media adherence revealed a statistically higher adherence when media had higher moisture content than when the media was dry. This was especially evident for the BSB sample, with

Table 3
Concentrations of elements across all media and particle size fractions.

		Concentration (μg/g)							
		Zn		As		Cd		Pb	
		gm	gsd	gm	gsd	gm	gsd	gm	gsd
CDA 1	<63 μm	3.11E+03	1.08	343	1.14	21.6	1.15	6.96E+03	1.07
	63–150 μm	3.83E+03	1.14	228	1.15	18.6	1.16	6.14E+03	1.13
	150–250 μm	2.09E+03	1.08	106	1.08	10.2	1.05	3.69E+03	1.08
	250 μm–2 mm	1.98E+03	1.31	67.8	1.25	23.3	1.51	3.82E+03	1.54
	>2 mm	1.05E+03	1.07	22.9	1.26	7.23	1.23	1.71E+03	1.81
CDA 2	<63 μm	1.16E+04	1.16	244	1.10	62.8	1.27	6.62E+03	1.11
	63–150 μm	7.56E+03	1.07	100	1.32	43.0	1.19	3.97E+03	1.08
	150–250 μm	2.47E+03	1.26	38.4	1.32	12.9	1.22	2.15E+03	1.23
	250 μm–2 mm	1.23E+03	1.42	20.6	1.38	5.55	1.39	1.43E+03	1.34
	>2 mm								
CDA 3	<63 μm	4.94E+03	1.03	297	1.14	43.1	1.09	6.26E+03	1.06
	63–150 μm	3.88E+03	1.03	147	1.04	26.5	1.04	4.46E+03	1.08
	150–250 μm	3.29E+03	1.06	108	1.17	29.6	1.12	5.74E+03	1.06
	250 μm–2 mm	4.58E+03	1.14	159	1.17	58.0	1.16	1.12E+04	1.12
	>2 mm	9.35E+03	1.19	426	1.37	98.5	1.51	8.51E+03	1.51
BSB	<63 μm	6.89E+03	1.03	87.4	1.06	11.5	1.05	740	1.04
	63–150 μm	2.04E+03	1.57	39.7	1.59	2.21	1.81	182	1.35
	150–250 μm	9.54E+03	1.04	45.0	1.13	1.86	1.58	383	1.36
	250 μm–2 mm	2.45E+04	1.07	31.1	1.20	1.00	1.00	554	1.25
	>2 mm	4.05E+02	1.66	9.28	2.24	2.47	2.28	201	2.77
Spokane River	<63 μm	2.91E+03	1.04	42.8	1.11	23.7	1.07	1.58E+03	1.08
	63–150 μm	3.13E+03	1.05	40.5	1.02	21.2	1.09	1.60E+03	1.10
	150–250 μm	3.14E+03	1.03	42.0	1.12	21.2	1.06	1.73E+03	1.05
	250 μm–2 mm	2.38E+03	1.11	31.0	1.06	17.2	1.11	1.66E+03	1.08
	>2 mm	650	1.28	10.2	1.05	2.45	1.26	303	1.21
IMQ 3/8 minus	<63 μm	135	1.07	76.4	1.07	0.200	1.00	27.1	1.22
	63–150 μm	87.1	1.11	38.7	1.07	0.200	1.00	20.0	1.00
	150–250 μm	95.9	1.24	29.2	1.05	0.186	1.15	16.8	1.41
	250 μm–2 mm	41.2	1.59	11.1	1.22	0.186	1.15	0.700	12.2
	>2 mm	69.3	1.18	20.2	1.37	0.200	1.00	13.2	1.73
IMQ 3/4–3/8	<63 μm	108	1.04	22.4	1.04	2.61	1.06	22.7	1.19
	63–150 μm	79.0	1.04	14.2	1.05	2.51	1.12	27.4	1.35
	150–250 μm	59.2	1.05	10.8	1.04	2.40	1.04	13.3	1.03
	250 μm–2 mm	63.3	1.17	6.95	1.10	2.69	1.11	6.92	1.12
	>2 mm	97.5	1.29	2.44	1.18	2.44	1.18	5.81	1.86
IMQ 3/4 minus	<63 μm	99.4	1.06	24.3	1.08	2.31	1.19	23.5	1.22
	63–150 μm	76.1	1.08	15.2	1.10	2.41	1.09	14.3	1.12
	150–250 μm	70.3	1.04	14.4	1.04	2.51	1.12	15.9	1.09
	250 μm–2 mm	58.2	1.40	8.67	1.31	2.32	1.12	2.32	1.12
	>2 mm	80.1	1.24	14.8	2.05	2.19	1.13	2.19	1.13
Everett slag	<63 μm	1.02E+04	1.04	2.04E+03	1.05	19.2	1.03	5.96E+03	1.06
	63–150 μm	6.97E+03	1.07	1.07E+03	1.09	11.7	1.08	3.72E+03	1.13
	150–250 μm	6.36E+03	1.10	734	1.05	8.15	1.20	2.98E+03	1.13
	250 μm–2 mm	2.23E+04	1.23	876	1.06	13.5	1.10	5.77E+03	1.17
	>2 mm	8.63E+04	1.10	788	1.13	30.4	1.26	2.94E+04	1.16
Reference soil 1a	<63 μm	57.3	1.41	9.74	1.05	0.186	1.15	22.1	1.22
	63–150 μm	33.9	2.24	3.35	1.69	0.186	1.15	14.1	2.00
	150–250 μm	22.7	1.29	1.12	1.49	0.168	1.41	0.433	4.69
	250 μm–2 mm	31.3	1.92	2.63	1.73	0.186	1.15	0.700	12.2
	>2 mm	145	2.70	5.79	1.57	0.186	1.15	31.3	3.81
Reference soil 1b	<63 μm	91.6	1.23	7.27	1.09	2.32	1.03	25.8	1.15
	63–150 μm	53.9	1.05	2.53	1.03	2.53	1.03	7.58	1.18
	150–250 μm	40.0	1.05	2.64	1.11	2.64	1.11	2.64	1.11
	250 μm–2 mm	45.2	1.05	2.10	1.26	2.10	1.26	2.70	1.22
	>2 mm	105	1.87	5.66	1.29	1.84	1.16	4.11	1.12

20.5 mg/cm^2 adhering in the wet trials and 0.0605 mg/cm^2 adhering in the dry trials. Differential adherence between wet and dry media is consistent with past findings (Kissel et al., 1996b).

3.4. Elemental mass adhering to hands

Total mass of elements adhering to hands in dry and wet trials are reported in Table 5. For all geologic media evaluated, higher elemental mass adhered to hands following wet trials than dry trials. In the Shapiro–Wilk test, normality for adhered elemental mass within gender and condition (dry or wet) could not be rejected for 166 of 176 datasets. Lognormality could not be rejected for 170 of 176 datasets. Therefore, lognormality was assumed for subsequent analyses. To determine if there was a difference in elemental mass adhering across genders, an independent sample t-test was performed using log transformed mass data. A significant difference was found in only 2 of 88 datasets ($p = 0.03$ and $p = 0.02$). Males and females were subsequently lumped and lognormality within condition (dry or wet) was tested using the Lilliefors modification of the Kolmogorov–Smirnov test. Lognormality could not be rejected in 77 of 88 datasets. Lastly, elemental adherence across wet and dry trials was tested using paired samples t-tests. A significant difference (mass adhering after wet trials higher than after dry trials) was found across all media for all elements (44 datasets). This is primarily due to higher media

Table 4
Summary of media loading on hands after dry and wet trials.

	Media loading (mg/cm^2)[a]			
	Dry trials		Wet trials	
	gm	gsd	gm	gsd
CDA1	1.14	1.20	4.71	1.44
CDA 2	0.475	1.27	12.2	1.63
CDA 3	0.655	1.27	1.80	1.75
BSB	0.0605	1.79	20.5	1.11
Spokane River	0.541	1.47	3.02	1.47
IMQ 3/8 minus	0.590	1.53	6.35	1.82
IMQ 3/4–3/8	0.191	2.10	0.892	1.37
IMQ 3/4 minus	0.255	1.63	8.53	1.70
Everett slag	0.694	1.42	9.50	1.75
Reference soil 1a	0.525	1.28	14.6	1.28
Reference soil 1b	0.500	1.42	7.40	1.52

[a] Geometric means and geometric standard deviations of six values collected during human subject trials for dry and wet media.

adherence rather than higher elemental concentrations in wet adhered media.

3.5. Wash water analysis

Wash water that passed through the filter was analyzed in an attempt to quantify any elemental mass that was not retained on filters. Maximum As, Cd, Pb, and Zn, percentages passing through the filter after dry handling trials were 18, 72, 8, and 20%, respectively. For As, Cd, and Pb, however, at least one of the concentrations in the water was below the detection limit. Filters used after wet media were handled resulted in lower elemental masses bypassing the filter than dry trials. Maximum percentage of element bypassing the filter for As, Cd, Pb, and Zn were 14, 42, 2, and 7%, respectively. For Cd and Pb, at least one of the concentrations in the water was below the detection limit. While it may appear that a large amount of Cd bypasses the filters (72 and 42% following dry and wet trials, respectively) there was a relatively small mass of Cd on the filters (<17.9 and <367 μg across all of the samples following dry and wet trials, respectively).

Overall, very little elemental mass bypassed the filters following the exposure scenario employed in this investigation. However, for all the elements discussed here, higher proportions of elements bypassed the filter following dry trials. This may reflect higher surface area to volume ratio rather than speciation or concentration differences. Generally, greater solubility implies greater bioavailability. The trials in which greater solubilization of elements appeared to occur suggest differences in relative bioavailability. However, formal solubility tests were not completed.

3.6. Comparison of bulk concentrations to adhered concentrations

Table 6 compares the reconstructed elemental concentrations observed in the bulk, unfractionated media to the elemental concentrations observed in the adhered media, after both wet and dry trials. For dry trials, As, Cd, Pb, and Zn had higher concentrations in adhered media in CDA-2, Spokane River, IMQ 3/8 minus, and Reference soil 1a. Likewise, for wet trials, As, Cd, Pb, and Zn had higher concentrations in adhered media than bulk media in the CDA-2 and Spokane River samples. Because As and Pb each have relatively large public health significance, they are of particular interest here. As and Pb concentrations in wet and dry adhered media are higher than concentrations in bulk media for CDA-1, CDA-2, Spokane River, IMQ 3/8 minus, IMQ 3/4–3/8, as well as the reference soils. This suggests preferential adherence of particle size fractions with higher elemental concentrations in many of the samples. Fig. 2 illustrate this effect as well. These figures compare wet and dry concentrations across the different media over the bulk media. Bars surpassing one indicate higher concentrations in adhered media. The trend discussed previously of higher concentrations in adhered media, however, was not observed in the Everett slag sample. In the Everett slag sample, the adhered media had lower concentrations of Cd, Pb, and Zn than the bulk media. This is most likely due to the high concentrations of those metals in the >2 mm fraction (Table 3) which did not appear to adhere to hands.

The above results (excluding those observed in the Everett slag sample) are similar to those found by Lepow et al. (1975) in their study of environmental lead exposure in urban children. The mean lead concentration on hands of children playing on residential soil was 2400 μg/g, compared to the mean level of 1200 μg/g in soil, suggesting preferential adherence of particles with high lead concentrations.

In the non-quarried and non-reference soils, there appear to be elevated levels of Pb, As, and Zn. Arsenic concentrations in

Table 5
Mass of elements adhering to hands following wet and dry handling trials.

Sample	Measure (μg)	Zn		As		Cd		Pb	
		Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet
CDA 1	gm	2.77E+03	1.33E+04	306	916	15.4	54.7	7.16E+03	2.55E+04
	gsd	1.13	1.37	1.14	1.39	1.20	1.40	1.20	1.38
CDA 2	gm	4.51E+03	7.08E+04	81.8	1177	17.9	367	3.66E+03	4.35E+04
	gsd	1.22	1.61	1.22	1.63	1.25	1.61	1.29	1.61
CDA 3	gm	2.63E+03	6.01E+03	171	307	16.8	40.6	3.65E+03	8.55E+03
	gsd	1.25	1.86	1.21	1.84	1.35	2.00	1.37	1.94
BSB	gm	983	3.79E+05	4.58	444	0.833	16.7	57.0	6.29E+03
	gsd	2.73	1.22	1.19	1.16	1.08	1.16	1.30	1.15
Spokane River	gm	1.50E+03	7.98E+03	25.2	127	8.87	50.0	769	4.60E+03
	gsd	1.41	1.67	1.36	1.67	1.43	1.68	1.44	1.66
IMQ 3/8 minus	gm	86.9	500	40.1	174	0.161	0.904	17.5	114
	gsd	1.44	1.53	1.55	1.59	1.46	1.76	1.66	1.80
IMQ 3/4–3/8	gm	28.3	64.2	2.69	8.10	6.14E−02	0.196	2.23	5.47
	gsd	1.81	1.30	2.19	1.31	1.53	1.76	1.73	1.60
IMQ 3/4 minus	gm	29.8	500	4.66	78.9	5.72E−02	1.19	3.24	39.5
	gsd	1.47	1.66	1.49	1.70	1.49	1.45	1.54	1.52
Everett slag	gm	5.74E+03	1.12E+05	1.38E+03	1.10E+04	9.01	111	3563	5.37E+04
	gsd	1.48	2.10	1.48	1.98	1.45	2.04	1.48	2.05
Reference soil 1a	gm	65.1	655	5.73	53.7	0.106	1.37	12.6	140
	gsd	1.25	1.33	1.39	1.23	1.52	1.76	1.53	1.56
Reference soil 1b	gm	56.2	318	4.51	21.4	0.104	1.30	12.4	37.3
	gsd	1.42	1.50	1.46	1.48	1.42	1.56	1.62	1.48

Table 6
Comparison of elemental concentrations across bulk media and adhered media across wet and dry trials.

	Zn (μg/g)			As (μg/g)			Cd (μg/g)			Pb (μg/g)		
	Bulk	Dry	Wet	Bulk	Dry	Wet	Bulk	Dry	Wet	Bulk	Dry	Wet
CDA 1	3.09E+03	2.57E+03	3.00E+03	199	284	206	16.8	14.3	12.3	5.38E+03	6.62E+03	5.72E+03
CDA 2	5.04E+03	1.01E+04	6.18E+03	77.2	183	103	27.8	40.0	32.0	3.14E+03	8.18E+03	3.78E+03
CDA 3	4.81E+03	4.26E+03	3.55E+03	219	277	181	43.5	27.2	24.0	6.37E+03	5.92E+03	5.04E+03
BSB	2.40E+04	1.72E+04	1.96E+04	31.4	80.4	22.9	1.04	14.6	0.855	548	1.00E+03	324
Spokane River	2.41E+03	2.94E+03	2.81E+03	32.1	49.5	44.5	16.8	17.4	17.6	1.46E+03	1.51E+03	1.61E+03
IMQ 3/8 minus	63.9	156	83.6	20.1	72.1	29.1	0.195	0.290	0.150	9.97	31.5	19.0
IMQ 3/4–3/8	97.1	154	74.6	2.67	14.5	9.41	2.45	0.333	0.227	6.08	12.1	6.35
IMQ 3/4 minus	78.6	121	60.7	14.5	19.0	9.58	2.21	0.233	0.144	3.10	13.2	4.80
Everett slag	5.73E+04	8.78E+03	1.25E+04	934	2.11E+03	1.23E+03	23.6	13.8	12.4	1.96E+04	5.45E+03	5.99E+03
Reference soil 1a	43.2	132	47.6	2.93	11.6	3.89	0.182	0.214	0.106	6.15	25.5	10.2
Reference soil 1b	50.0	117	44.6	2.69	9.35	3.00	2.27	0.215	0.182	4.13	25.7	5.23

uncontaminated soils usually range from 1 to 40 μg/g (Mandal and Suzuki, 2002). Arsenic in Washington State's soil varies between 1 and 10 μg/g (WA State DOE, 1994). Arsenic in the Everett slag sample under investigation here had a mean concentration of 934 μg/g in bulk media. For comparison, in a study conducted by Polissar et al. (1990), the mean As soil concentration obtained in Ruston, WA households with a median distance of 0.3 miles from a copper smelter was 353 μg/g.

In addition, several of the geologic media under study here (Table 6) have noticeably high lead concentration levels. For example, the Everett slag sample has a bulk soil lead concentration of 19,600 μg/g. Roels et al. (1980) observed elevated soil lead concentrations from 2000 to 6000 μg/g within a distance of 1 km from a lead smelter that had produced approximately 100,000 metric tons of lead per year. The Everett slag sample bulk media lead concentration well exceeds those observed by Roels et al. as well as the natural lead concentration of soil obtained from crustal rock which ranges from <10 to 30 μg/g Agency for Toxic Substances and Disease Registry (ATSDR) (2007). In Washington State, lead concentration tends to be between 2 and 20 μg/g, with higher concentrations in more densely populated regions (most likely due to automobile exhaust) (WA State DOE, 1994). As revealed by the media investigated here, the top layers of soil may vary greatly in lead concentration due to industrial activities occurring in the vicinity.

Zinc concentrations in the media analyzed in this study also appear to be elevated compared to background levels. The highest mean

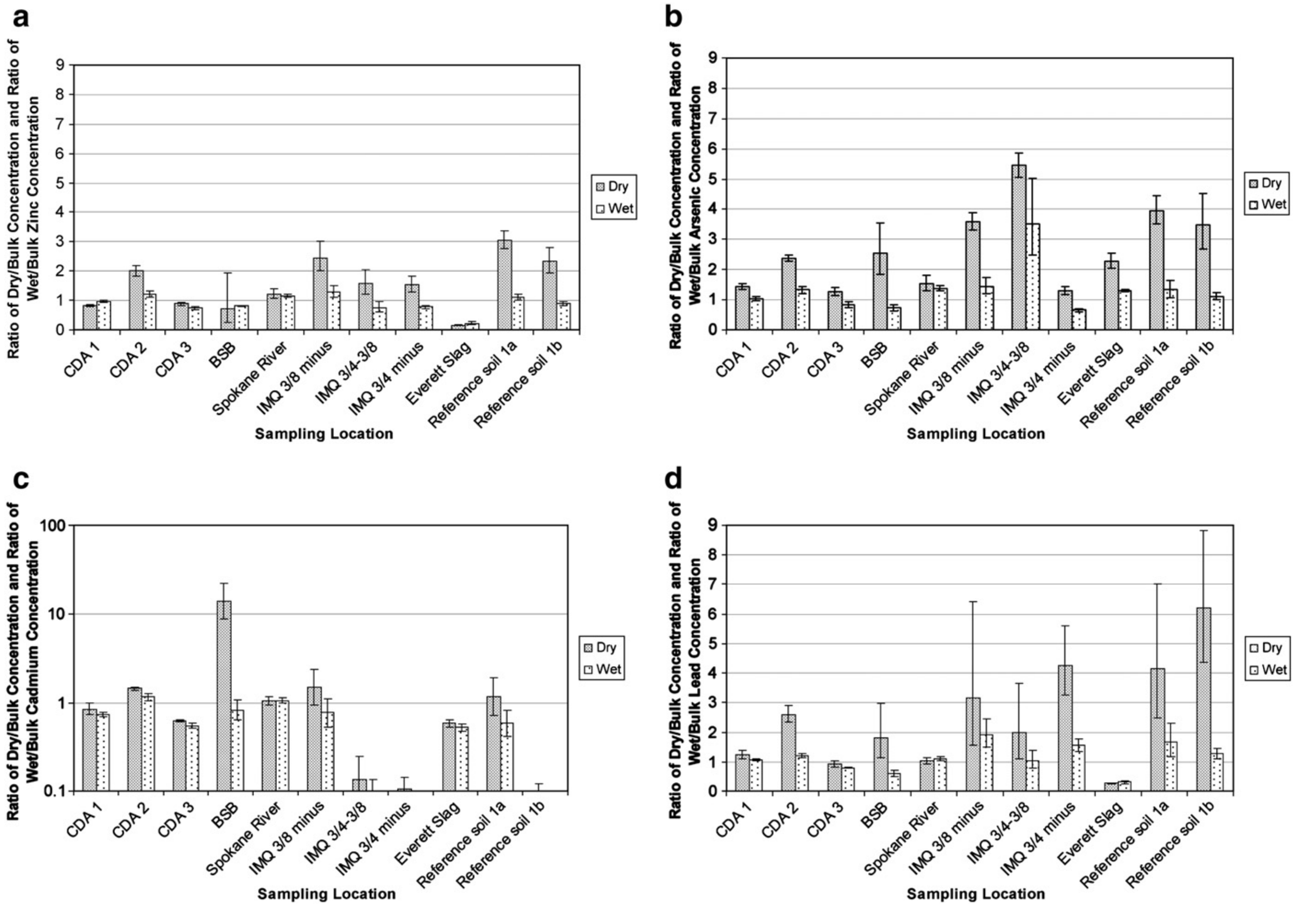


Fig. 2. a–d. Ratio of dry/bulk concentrations and wet/bulk concentrations in media for zinc, arsenic, cadmium, and lead, respectively. Note that scaling for cadmium in (Fig. 2c) is different than the other elements. Columns represent geometric means. Error bars are 95% confidence intervals.

Table 7
MLE results for dry adhered media.

	Particle size	Predicted fractions[a]		Particle size	Predicted fractions[a]
CDA 1	<63 µm	0.763	IMQ 3/8 minus	<63 µm	1.000
	63–150 µm	0.000		63–150 µm	0.000
	>150 µm	0.237		>150 µm	0.000
CDA 2	<63 µm	0.632	IMQ 3/4–3/8	<63 µm	0.604
	63–150 µm	0.239		63–150 µm	0.000
	>150 µm	0.129		>150 µm	0.396
CDA 3	<63 µm	0.808	IMQ 3/4 minus	<63 µm	0.457
	63–150 µm	0.192		63–150 µm	0.0904
	>150 µm	0.000		>150 µm	0.453
BSB	<63 µm	0.875	Everett slag	<63 µm	0.770
	63–150 µm	0.000		63–150 µm	0.230
	>150 µm	0.125		>150 µm	0.000
Spokane River	<63 µm	0.000	Reference soil 1a	<63 µm	1.000
	63–150 µm	0.000		63–150 µm	0.000
	>150 µm	0.214		>150 µm	0.000
			Reference soil 1b	<63 µm	1.000
				63–150 µm	0.000
				>150 µm	0.000

[a] Seed values were 0.33 across all particle fractions and across all samples.

concentration of Zn in this investigation was 57,500 µg/g in the Everett slag bulk media. In the conterminous United States, the concentration of Zn in soils ranges between <5 and 2900 µg/g, with a mean of 60 µg/g at a depth of 20 cm from the surface (Shacklette and Boerngen, 1984). In Washington State, Zn concentrations vary between 10 and 100 µg/g (WA State DOE, 1994). In eastern Pennsylvania, the concentration of zinc in soil at the Palmerton zinc smelter was measured 6 years after termination of the zinc smelting operation in 1980 (Storm et al., 1994). The highest level of Zn found was 4160 µg/g at the sampling location closest to the smelter. Zinc concentrations decreased with distance from the smelter (Storm et al., 1994). Based on the above data, elevated zinc concentrations are present in many of the sampling locations of this study.

3.7. Maximum Likelihood Estimation (MLE) results

Tables 7 and 8 display the MLE results conducted across all media for dry and wet trials, respectively. Greater than 60% of the adhered fraction following dry trials was estimated to be <63 µm in many of the samples including CDA-1, -2, -3 and BSB. The two samples where this was not observed were in Spokane River and IMQ 3/4 minus. Following wet trials, it was estimated that the <63 µm fraction accounts for less than 25% in many of the samples including CDA-1, -2, -3, as well as the Everett slag sample.

The results presented here also indicate that whole, unfractionated media elemental concentrations may not reflect actual concentrations adhering to hands. Smaller particle size fractions appear to adhere to hands, especially when the media is dry. This is evidenced by the MLE exercise and supports the findings of prior investigators (Choate et al., 2006; Driver et al., 1989; Kissel et al., 1996b; Sheppard and Evenden, 1994; Yamamoto et al., 2006). In addition, the regression analyses indicate that smaller particle sizes tend to have higher elemental concentrations. This may lead to an overall underestimation of potential exposures to the soil contaminants studied here if elemental concentrations in bulk media were used in an exposure assessment.

3.8. IEUBK Model

For the IEUBK modeling exercise, blood lead levels were estimated for two to five year olds as a result of ingestion of lead-contaminated soil. Absorption of lead from dust ingestion, water ingestion, and air inhalation was assumed to be zero to isolate the soil ingestion route. All parameters except soil lead concentration were default EPA values. This included a 30% gastrointestinal absorption rate and soil ingestion rates for 2–3, 3–4, and 4–5 year olds of 0.135, 0.135, and 0.100 g/day, respectively. Ingestion rates for 0–1, 1–2, 5–6, and 6–7 year olds were set at 0 g/day because this exercise concentrated on lead exposure to 2–5 year olds, an age range which often exhibits the largest amount of

Table 8
MLE results for wet adhered media.

	Particle size	Predicted fractions[a]		Particle size	Predicted fractions[a]
CDA 1	<63 µm	0.234	IMQ 3/8 minus	<63 µm	0.000
	63–150 µm	0.398		63–150 µm	0.867
	>150 µm	0.368		>150 µm	0.133
CDA 2	<63 µm	0.206	IMQ 3/4–3/8	<63 µm	0.346
	63–150 µm	0.376		63–150 µm	0.000
	>150 µm	0.418		>150 µm	0.654
CDA 3	<63 µm	0.227	IMQ 3/4 minus	<63 µm	0.000
	63–150 µm	0.773		63–150 µm	0.187
	>150 µm	0.000		>150 µm	0.813
BSB	<63 µm	0.000	Everett slag	<63 µm	0.188
	63–150 µm	0.207		63–150 µm	0.729
	>150 µm	0.793		>150 µm	0.0825
Spokane River	<63 µm	0.000	Reference soil 1a	<63 µm	0.147
	63–150 µm	0.640		63–150 µm	0.316
	>150 µm	0.360		>150 µm	0.537
			Reference soil 1b	<63 µm	0.106
				63–150 µm	0.124
				>150 µm	0.770

[a] Seed values were 0.33 across all particle fractions and across all samples.

Table 9
Results of IEUBK simulation using concentrations obtained from handling trials and reconstructed bulk concentrations for 2–5 year olds. Highlighted cells exceed 10 μg/dL.

	Geometric mean blood lead level: 2–5 year olds (μg/dL)		
	Bulk	Dry	Wet
CDA 1	26.1	29.5	27.1
CDA 2	18.5	33.4	20.9
CDA 3	28.9	27.6	25.1
BSB	4.61	7.76	2.85
Spokane River	10.5	10.8	11.4
IMQ 3/8 minus	0.115	0.314	0.198
IMQ 3/4–3/8	0.0790	0.134	0.0810
IMQ 3/4 minus	0.0510	0.145	0.0670
Everett slag	54.8	26.3	27.8
Reference soil 1a	0.0790	0.259	0.117
Reference soil 1b	0.0610	0.260	0.0710

hand-to-mouth behavior and unintentional soil ingestion. The soil/dust weight factor was set at 75% soil (i.e., the model runs completed here assume that 75% of the soil and dust ingested is soil). Table 9 summarizes the results from the model runs completed using bulk, dry-adhered, and wet-adhered lead concentrations in all 11 media for 2–5 year olds. In 5 of the 11 samples, the estimated blood lead level exceeded the CDC's health effects limit of 10 μg/dL (CDC, 1991). The highest blood lead level estimated was 54.8 μg/dL (using the bulk Everett slag media lead concentration).

Comparison of blood lead levels predicted to result from ingesting bulk media or adhered media reveals that for 5 of the 11 samples, the estimated blood lead levels exceed 10 μg/dL. Results for CDA-2 and Everett slag display the largest difference in blood lead levels estimated when using bulk concentrations versus using concentrations in adhered media (of those media with estimated blood lead levels above 10 μg/dL). For CDA-2, use of the lead concentration in bulk media may lead to underestimation of actual blood lead level. Conversely, the blood lead level estimated using lead concentration in bulk media is higher than that produced by use of adhered lead concentration for Everett slag. This is due to the elevated lead concentrations in the >2 mm fraction, which actually were slag and which did not appear to adhere, as discussed earlier. The fines in the Everett slag sample included non-slag material with much lower As and Pb levels.

Because the IEUBK Model assumes that the soil concentrations occur in residential yards, or areas where children spend a majority of their time, and materials studied here (except the reference soil) are not yard soils, values reported in Table 9 may be overestimates. However, results in Table 9 reflect the effect of concentration only. Whether exposure to wet soils leads to greater adherence and hence greater rates of soil ingestion sufficient to compensate for lower soil concentration is not known. Some of the media investigated here were collected in close proximity to bodies of water, and might be encountered wet or by wet skin. On the other hand, the duration that wet loads on skin stay wet and/or stay on skin is also not known. As wet loads dry, some sloughing could be expected.

4. Conclusions

The media investigated here are derived from mining, smelting and quarrying activities and are not conventional soils. Many exhibit elevated levels of arsenic and lead. Nevertheless, some results are consistent with prior reports for soils. Higher media loading was observed when media were wet, and the smallest particle sizes appeared to adhere preferentially following contact with dry as opposed to wet media. Modest particle size-concentration effects (usually manifest as higher elemental concentrations in smaller particles) also appear to occur in most of the media investigated. As a consequence, media adhering dry generally were found to have higher As, Cd, Pb, and Zn concentrations than bulk media. With respect to Pb, the potential effects of differences in concentrations in bulk media and media adhering dry (Table 6) can be seen in IEUBK predictions in Table 9. These outcomes are generally consistent with the view that use of elemental concentrations in bulk media in exposure/risk assessment may lead to underestimation of actual exposures to children having access to the sampled media and exhibiting hand-to-mouth behavior. In one unusual case (Everett slag), however, very high lead levels in the coarsest fraction resulted in lower concentrations in adhering media (wet or dry) than in bulk media. Nevertheless persons conducting sampling of soil and soil-like materials for purposes of risk assessment should consider soil fractionation strategies carefully in light of recent research regarding adhering particle size. Lastly, while blood lead levels estimated here may be overestimates of potentially affected children's blood lead levels since the target media are not residential soils, the results do suggest that some of the target media present non-negligible risk of elevated lead exposure to children.

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