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# A multi-element profile of housedust in relation to exterior dust and soils in the city of Ottawa, Canada

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## Abstract

This paper presents multi-element profiles of indoor dust versus exterior soils and dusts from 50 residences located in 10 neighborhoods across Ottawa, the capital city of Canada. Mercury concentrations were determined using nitric–sulphuric acid digestion and cold vapor AAS. Concentrations of 31 other elements were determined using nitric–hydrofluoric acid digestion and ICP-MS. Comparisons of household dust, garden soil and street dust at the individual residence scale and at the community scale were based on a consistent 100–250- $\mu\text{m}$  particle size fraction. Results showed housedust samples to contain significantly higher concentrations of many key elements, including lead, cadmium, antimony and mercury, than either street dust or garden soil samples. Also, housedust profiles revealed a distinct multi-element signature in relation to exterior dust and soil samples. Interestingly, garden soil contained higher concentrations of aluminum, barium and thallium than either house or street dust. Geometric mean concentrations (mg/kg) of these elements in household dust/garden soil were: lead 233/42; cadmium 4.42/0.27; antimony 5.54/0.25; mercury 1.728/0.055; aluminum 24281/55677; barium 454/763; and thallium 0.14/0.29. Street dust contained lower geometric mean concentrations than garden soil for 23 out of a total of 32 elements. In general, indoor/outdoor concentration ratios varied widely from one element to another, and from one residence to another within the community. In the case of Ottawa, which is a city with a low concentration of heavy industries, it would be difficult-to-impossible to accurately predict indoor dust concentrations based on exterior soil data. It is concluded that dust generated from sources within the house itself can contribute significantly to exposures to certain elements, such as lead, cadmium, antimony and mercury. © 2001 Elsevier Science B.V. All rights reserved.

*Keywords:* House dust; Street dust; Soil; Metal sources; Lead exposure; Risk assessment; Inductively coupled plasma mass spectrometry; Cold vapor atomic absorption spectrometry; Trace elements

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## 1. Introduction

Ingestion of dust and soil is widely regarded as the key pathway for childhood exposure to lead-based paint, leaded gasoline, and other metals and metalloids derived from vehicular traffic and local industrial sources (Duggan and Inskip, 1985; HWC, 1992; Thornton et al., 1994; Gulson et al., 1995; Lanphear et al., 1998; Mielke and Reagan, 1998; Matte, 1999; Meyer et al., 1999). Many studies have treated ingestion of household dust and soil as a single exposure pathway, a practice which can lead to erroneous conclusions (Gulson et al., 1995). Recently White et al. (1998) emphasized the need to improve residential exposure assessments by disaggregating dirt ingestion into separate categories for indoor housedust and exterior dirt. Currently however, there is a scarcity of data that distinguish indoor dust from exterior soil (White et al., 1998). Such data are needed to more accurately determine exposures of preschoolers, especially older infants and toddlers, who spend most of their time indoors and ingest dust through normal repetitive hand-to-mouth activities (Duggan and Inskip, 1985; White et al., 1998; Mushak, 1998). Urban survey data indicate wide variations in metal concentrations of dust and soil in different activity areas within a residence, amongst different residences within a community, and amongst different communities (Elhelu et al. 1995; Gulson et al., 1995; Sutton et al., 1995; de Miguel et al., 1997; Meyer et al., 1999), underscoring the need for more representative, site-specific data to improve residential exposure assessments. A key question is the degree to which the metal content of housedust originates from the transport of outdoor dusts and soil into the interior of a home. Estimates of the contribution of soil lead to housedust lead range from 20 to 95%, depending on a wide variety of site-specific factors and methodological approaches (Gulson et al., 1995; Mielke and Reagan, 1998; Meyer et al., 1999 and references cited therein).

The present study was undertaken to character-

ize total concentrations of a broad spectrum of major, minor and trace elements in urban dusts representative of Ottawa, the capital city of Canada, and to compare the multi-element profile of indoor dusts to that of exterior dusts and soils. Ottawa is located in south-eastern Ontario, with a population of 323 340 and land area of 110 km<sup>2</sup> (Statistics Canada, 1996). Many Canadian urban dust studies have focused on lead, particularly in communities with industrial 'hotspots', such as the south Riverdale neighborhood in Toronto, Ontario, and Trail, British Columbia (Stokes, 1988; Health Canada, 1994). In contrast, Ottawa has a low concentration of heavy industries. The largest employers are government, health services, communications and high technology firms, three universities and two colleges. Most of the phase-out of lead additives from gasoline occurred in the mid- to late-1980s, and was finalized in 1990 (by regulation). In a previous study of soil lead concentrations in urban Ottawa, Ericson and Mishra (1990) reported a range of 7.5–100 mg/kg (geometric mean 28.4 mg/kg) for surface soil samples collected from 26 sites. This range is low in comparison to soil lead concentrations reported for other North American cities, which are commonly higher than 1000 mg/kg (Stokes, 1988; Rabinowitz, 1992; Elhelu et al., 1995; Sutton et al., 1995). The purpose of the present study is to obtain baseline data, not only for lead, but also for a number of other elements, so that the relative importance of indoor and outdoor sources of exposure may be determined and profiled precisely. Toward this goal, total element concentrations were determined in a consistent particle size fraction (100–250- $\mu$ m) of household vacuum cleaner dusts, garden soil and street dust samples from 50 residences located in 10 different zones in Ottawa. Such a survey design should provide a measure of the variability amongst interior and exterior media within an individual residence, and amongst different residences at the community scale, and thus should provide a more representative multi-element signature for this non-industrial North American urban setting.

## 2. Methods

### 2.1. Sample collection

House-dust samples were collected from a total of 50 residences selected at random from 10 different zones of Ottawa. In addition, street dust samples and garden soil samples were collected in the immediate vicinity (within 15 m) of each residence. The survey was conducted in the winter of 1993 by Concord Environmental Corporation, Gloucester, Ontario. Each participating householder completed a questionnaire to document the: age; size; construction characteristics; heating sources; renovation/redecoration history of the house; distance from the road; number of adult occupants, children and pets; and lifestyle factors including occupations and smoking habits.

The residences surveyed in this study were private dwelling units built between 1893 and 1987. Out of the total of 50 dwellings, 28% (14) were built prior to 1946. The Statistics Canada 1991 Census indicates that 18.6% (25 640) of the total number of private dwelling units in Ottawa (138 065) were built prior to 1946. Thus, this survey includes a somewhat higher proportion of older dwelling units than does the city as a whole. At the time of the survey (1993) the median house age was 40 years. Gas furnaces were used as the primary heat source in 27 residences, including the two oldest houses built in the 1800s, while 14 used oil furnaces and seven used electricity as the primary heat source. The median age (in 1993) of gas-heated houses was 41 years, compared to 40 years for oil-heated houses and 18 years for electrically heated houses.

Residents themselves collected the indoor dusts by vacuuming according to a protocol designed to capture recent surface dusts and avoid longer-term sinks of historical materials. New vacuum cleaner bags were provided for the sample collection, and participants were instructed to vacuum the house 1 week prior to the sampling, and to record the dimensions and characteristics of the vacuumed surfaces, which included bare floors,

area rugs and broadloom. Following sampling, vacuum cleaner bags were sealed with paper by Concord personnel and then resealed inside a plastic bag for shipment to the Health Canada laboratory in Ottawa. Street-dust samples were swept using a clean plastic brush and dust pan, from five separate 1 m<sup>2</sup> sections of street directly adjacent to each residential property, typically 5–10 m from the house itself. A clean stainless steel trowel was used to collect and composite the top 5 cm of garden soil from five locations in the yard of each household. After removal of larger pieces of debris (for example, leaves, twigs and wood chips, and in the case of the street dusts, bottle caps and cigarette butts), the exterior samples were double bagged in clean polyethylene bags for shipment to the laboratory.

### 2.2. Sample preparation

The samples were oven-dried at 90°C for 6 h, sieved through nylon mesh, and stored in glass vials at ambient temperature by Health Canada (Environmental Health Directorate) laboratory personnel. Measurement of a consistent particle size fraction was a priority consideration, as finer size fractions generally have higher element concentration values than coarser fractions or bulk samples (Duggan and Inskip, 1985; Gulson et al., 1995). In this study, the 100–250- $\mu\text{m}$  particle size fraction was used for analysis, with the exception of some of the street dust samples for which it was necessary to add the 75–100- $\mu\text{m}$  fraction to yield an adequate sample mass. Out of the 50 houses surveyed, five street dust samples and two housedust samples were inadequate for subsequent analysis, and in their place, standard reference materials (SRMs) were submitted 'blind' to the analytical laboratory (discussed below). In total, the final submission consisted of a total of 45 street dust samples, 48 housedust samples, 50 garden soil samples, and 10 blind standards.

### 2.3. Analytical procedure

Analytical work was performed by RPC Chemical and Biotechnical Services, Fredericton, New

Brunswick, Canada using inductively coupled plasma mass spectrometry (ICP-MS) for multi-element determinations and cold-vapor atomic absorption spectrometry (AAS) for mercury determinations.

### 2.3.1. Total mercury determination by cold vapor AAS

The digestion method for total mercury determination in soils and dusts consisted of heating 400 mg of the sample in a mixture of high purity nitric acid (2 ml) and sulfuric acid (5 ml) in a water bath at 80°C for 2 h. After cooling, excess 5% potassium permanganate (6 ml) and 5% potassium persulfate (2 ml) were added to the digests, which were allowed to sit overnight to complete the oxidation of organic matter. The next day hydroxylamine hydrochloride (20% solution) was added dropwise to reduce permanganate and manganese oxides, and mercury was determined using stannous chloride reduction and cold vapor AAS detection.

### 2.3.2. Multi-element determination by ICP-MS

To digest soil and housedust samples for ICP-MS analysis, a 250-mg sample was placed in a mixture of high purity nitric acid (2.5 ml) and hydrofluoric acid (2.5 ml) and allowed to sit overnight at ambient temperature. Street dusts required the addition of perchloric acid (0.4 ml) at this stage to dissolve an acid-resistant organic component, thought to be asphalt. After slow evaporation to dryness, nitric acid was added (1 ml) and the solutions were evaporated again, followed by a final addition of nitric acid (2 ml) and hydrochloric acid. Hydrochloric acid volume was kept to a minimum (10 drops) to reduce molecular ion interferences in the ICP-MS determination. Water was added to provide an effective dilution of 200:1 and the solutions were then heated at 80°C to re-dissolve the metals. Additional dilutions were applied as necessary prior to analysis. Multi-element analyses were performed using a Fisons/VG Plasma Quad II STF instrument equipped with a 27.12-MHz radio frequency generator, a Gilson Minipuls-3 peristaltic pump, a Meinhard concentric glass nebulizer, and a water-jacketed double-pass quartz spray chamber.

The instrument was operated in the 'dual detector' mode with data acquisition by 'peak jump' for optimum precision. RF power was maintained at 1350 W incident and 0–1 W reflected. The plasma gas flow rate was 12 l/min and the auxiliary gas flow rate was 0.95 l/min. Nickel sampler and skimmer cones were used for all analyses. The MS vacuum pressure operated in the range of  $1.1 \times 10^{-6}$ – $2.5 \times 10^{-6}$  mbar. The sample delivery rate was approximately 2 ml/min. The total data acquisition time was 60 s/sample, and the uptake and rinse-out times were 90 s/sample. A separate 2% nitric acid wash was used between samples (10 s). Internal standards (Rh and Th) were used to correct for variations in plasma conditions, detector drift and nebulization efficiency. A series of eight multi-element standards, containing from two to 30 elements each, were used to calibrate the instrument over the operating range. Each sample batch prepared for ICP-MS analysis included samples, duplicates, blanks and standard reference materials. Inductively coupled plasma-optical emission spectroscopy (ICP-OES) was used to confirm that concentrations of the major elements were in the linear response range for the ICP-MS.

### 2.3.3. Quality control and quality assessment

Each sample batch prepared for ICP-MS and cold-vapor AAS analyses included sample duplicates, blanks and certified standards. There were a total of 22 sample duplicates: 6 duplicates of housedust, 4 duplicates of garden soil, and 10 duplicates of street dust. In addition, two housedust samples were split and submitted separately as field duplicates for an assessment of sample heterogeneity. Blind standards (10) were submitted (as indicated above) and the RPC laboratory included an additional 14 standards for ICP-MS determination and 6 standards for cold-vapor AAS determinations. The standards analyzed in this study were NRC marine sediment standards MESS-1 (multi-element,  $n = 7$ ) and BEST-1 (mercury only,  $n = 6$ ); NIST 1648 (urban particulate matter,  $n = 3$ ); NIST 2704 (Buffalo River sediment,  $n = 7$ ); NIST 2709 (San Joaquin soil,  $n = 1$ ); NIST 2710 (Montana I soil,  $n = 1$ ); NIST 2711 (Montana II soil,  $n = 1$ ); CANMET CCRMP

soil standards SO-2 (Podzolic B horizon soil,  $n = 1$ ), SO-3 (Calcareous C horizon soil,  $n = 1$ ), and SO-4 (Chernozemic A horizon soil,  $n = 1$ ).

### 3. Results

#### 3.1. Analytical parameters

Recovery and analytical reproducibility in Table

1a (ICP-MS) and Table 1b (cold-vapor AAS) were evaluated using standards and sample replicates. Analyses of 10 different standard reference materials (SRMs) allowed an assessment of accuracy and precision over a wide range of element concentrations. Concentration data for lead, mercury, arsenic, and cadmium determinations of SRMs plotted against certified values (Fig. 1) indicate that observed values correlate well with certified values over two or more orders of magni-

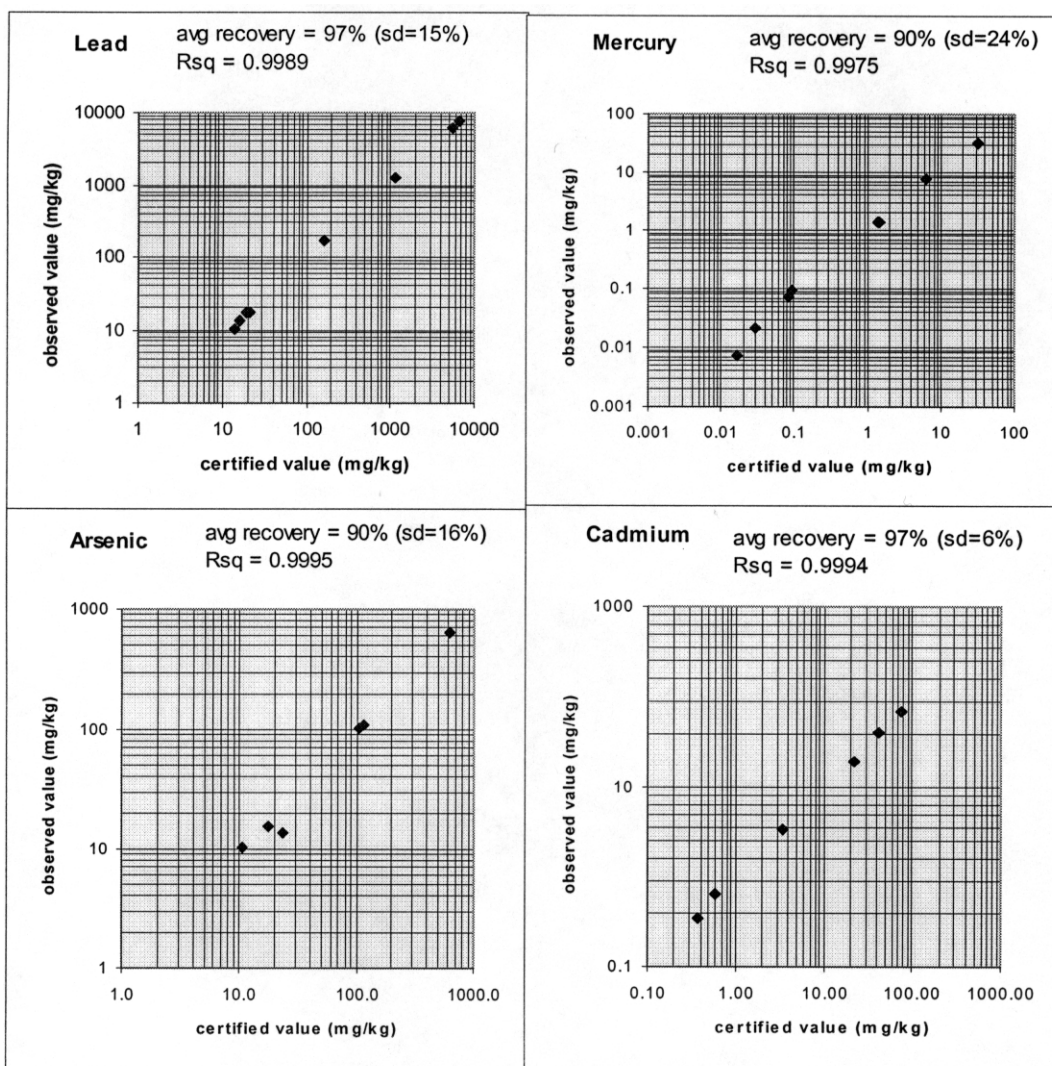


Fig. 1. Concentration data for lead (Pb), mercury (Hg), arsenic (As), and cadmium (Cd) determinations of standard reference materials plotted against certified values. Determinations by ICP-MS for Pb, As, and Cd and by cold vapor AAS for Hg. ‘Rsq’ =  $R^2$  values were calculated for linear correlations. Average recoveries are based on all of the SRM recovery data obtained in this study, including those in Table 1a and Table 1b.

tude. Recoveries for total lead averaged 97% ( $\pm 15\%$ ) over three orders of magnitude in nine different SRMs (Fig. 1). Based on NIST 2709, 2710, and 2711 (not shown), recoveries for silver averaged 100% ( $\pm 15\%$ ) and recoveries for molybdenum averaged 106% ( $\pm 4\%$ ). Tables 1a,b indicate quantitative recoveries for the other elements, with the exception of chromium in NIST 1648 (certified value  $403 \pm 12$  mg/kg Cr). Chromium recoveries were acceptable (86% average) in the six other SRMs which have lower chromium concentrations, more representative of the range encountered in this study, with a strong correlation between observed and certified values ( $R^2 = 0.98$ ;  $n = 6$ ).

Analytical reproducibility was better than 10% relative standard deviation (RSD) for most of the samples (Table 1a,b). There were, however, notable exceptions for certain elements, particularly bismuth, selenium and tellurium, which occur in concentrations close to the minimum detectable limit (MDL; listed in Tables 2–4). For most elements, median values were more than 10-fold greater than the MDL. Precision for garden soil duplicates was typically better than precision for housedust and street dust duplicates (see RSD values in Table 1a,b), suggesting that sample heterogeneity contributes to higher variation in the latter two sample types. Flakes of paint were observed during microscopic examination of selected samples, and are a potential source of dust sample heterogeneity on the 0.25-g scale used for these analyses. Other potential sources of heterogeneity include nodules of solder, mercury from broken thermometers, scale, corrosion products, and metal filings.

### 3.2. Total element concentrations and element associations

A comparison of the total elemental concentrations summarized in Tables 2–4 indicates that housedust samples contain significantly higher concentrations of many elements, including lead, cadmium, antimony and mercury, than either street dust or garden soil samples. Thus, the geometric mean lead concentration in housedust

(Table 3) is over five times higher than that of garden soil (Table 2). In fact, concentrations of lead in housedust samples exceed street dust lead concentrations in all 48 residences, and exceed garden soil lead concentrations in all but one residence. Similarly, mercury concentrations in housedust exceed street dust and garden soil mercury concentrations in 47 out of 48 homes sampled. House-dust cadmium concentrations exceed street dust and garden soil cadmium concentrations in all 48 residences. For a number of other elements, however, the opposite trend is observed. Garden soil contains higher concentrations of aluminum, barium and thallium than either housedust or street dust (Tables 2–4). Street dust contains lower geometric mean concentrations than garden soil for 23 out of a total of 32 elements (Tables 2 and 4), an observation which may be attributed variously to losses of soluble species from street dust by run-off during municipal street cleaning, greater attenuation in garden soils due to the higher organic matter content of the soil matrix, wash-off of settled dust, paint chips, other particulate matter from houses, and leaching of soluble species from exterior wall and roof materials.

Some significant elemental associations in the housedust samples ( $n = 48$ ) may indicate common indoor sources. Linear regression analyses (at 95% confidence level) show positive correlations between aluminum and a number of elements in housedust, including beryllium ( $R^2 = 0.89$ ), potassium ( $R^2 = 0.79$ ), vanadium ( $R^2 = 0.76$ ), rubidium ( $R^2 = 0.75$ ), and strontium ( $R^2 = 0.64$ ). Thallium also correlates with beryllium ( $R^2 = 0.56$ ) and rubidium ( $R^2 = 0.59$ ), and more weakly with aluminum ( $R^2 = 0.49$ ). Positive correlations also occur between barium and cobalt ( $R^2 = 0.53$ ) in housedust, and there is a positive but weak association between lead and barium ( $R^2 = 0.35$ ), and between lead and cobalt ( $R^2 = 0.28$ ). It is notable that these correlations are not observed in either the garden soil or street dust data sets, which have distinctly different elemental associations. No significant correlations were observed in comparisons of element concentrations in housedust versus street dust, or housedust versus garden soil.

Table 1a

Recovery and reproducibility based on samples and standard reference materials for ICP-MS multi-element analyses

Element	Sample replicates			Standard replicates								
	House dust (6 pairs) (% RSD)	Garden soil (4 pairs) (% RSD)	Road dust (10 pairs) (% RSD)	MESS-1 ( <i>n</i> = 7)			NIST 2704 ( <i>n</i> = 7)			NIST 1648 ( <i>n</i> = 4)		
				Observed <sup>a</sup> (mg/kg)	RSD <sup>b</sup> (%)	Recovery <sup>c</sup> (%)	Observed mg/kg	RSD %	Recovery %	Observed mg/kg	RSD %	Recovery %
Aluminum	18	3	8	56986	7	98	59186	8	97	30733	6	90
Antimony	19	6	19	0.71	14	98	3.7	4	97			
Arsenic	9	4	52	10.2	4	96	13.5	22	58	111.1	15	97
Barium	11	1	4				401	6	97			
Beryllium	14	3	8	2.02	6	106						
Bismuth	14	35	37									
Cadmium	29	17	27	0.64	3	108	3.4	2	98	71.4	1	95
Calcium	5	2	5	4830	4	100	25843	2	99			
Chromium	7	3	9	55.3	6	78	133.2	6	99	112.7	5	28
Cobalt	6	2	5	10.69	4	99	14.07	2	100			
Copper	37	9	29	28.91	9	115	104.0	7	105	580.1	7	95
Iron	11	4	8	29800	6	98	39414	5	96	34600	5	88
Lead	23	4	21	32.19	7	95	173.5	12	108	7593.4	25	116
Lithium	11	4	7				45.2	8	95			
Magnesium	7	2	7	8646	5	99	12229	4	102			
Manganese	10	3	8	485.7	9	95	559.1	6	101			
Molybdenum	15	8	12									
Nickel	15	1	9	29.1	4	99	48.7	16	110	78.5	4	96
Phosphorus	7	11	12	633	8	99	941	6	94			
Potassium	8	1	5	18657	5	100	19443	8	97	9467	6	90
Rubidium	8	2	5									
Selenium	26	8	45	Below detection			1.4	13	129	23.8	12	88
Silver	54	18	14									
Sodium	6	2	6	17643	8	95	5336	10	98	3720	14	88
Strontium	14	2	9									
Tellurium	38	141	37									
Thallium	10	5	5				1.04	4	98			
Tin	32	8	36	3.85	10	97						
Uranium	9	30	13				2.48	18	79	5.66	21	103
Vanadium	15	13	7	71.6	6	99	92.1	7	97	128.0	2	91
Zinc	13	4	13	191.1	5	100	436.6	4	100	44477.5	5	94

<sup>a</sup>‘Observed’ value is the mean concentration in mg/kg.<sup>b</sup>‘RSD’ is relative standard deviation = (std.dev./mean × 100).<sup>c</sup>Recovery = (observed value/certified value × 100); observed values for samples varied over the concentration ranges in this study (Tables 2–4).

Table 1b

Recovery and reproducibility based on samples and standard reference materials for cold vapor AAS determination of mercury

Sample replicates			Standard replicates					
House dust (6 pairs) (% RSD)	Garden soil (4 pairs) (% RSD)	Road dust (10 pairs) (% RSD)	BEST-1 ( <i>n</i> = 6)			NIST 2704 ( <i>n</i> = 7)		
			Observed <sup>a</sup> (mg/kg)	RSD <sup>b</sup> (%)	Recovery <sup>c</sup> (%)	Observed (mg/kg)	RSD (%)	Recovery (%)
24	12	20	0.096	10	106	1.416	9	96

<sup>a</sup>‘Observed’ value is the mean concentration in mg/kg.<sup>b</sup>‘RSD’ is relative standard deviation = (std.dev./mean × 100).<sup>c</sup>Recovery = (observed value/certified value × 100); observed values for samples varied over the concentration ranges in this study (Tables 2–4).

Table 2

Total element concentrations (mg/kg) in garden soil of Ottawa residences (*n* = 50)

Element <sup>a</sup>	MDL <sup>b</sup> (mg/kg)	Soil Quality Criteria <sup>c</sup>	Arithmetic mean	Geometric Mean	Median	Minimum Value	Maximum value	90th percentile	95th percentile
Aluminum	5	–	55841	55677	55750	45400	64200	61730	62220
Antimony	0.02	13	0.36	0.25	0.22	0.11	1.98	0.77	1.00
Arsenic	0.5	(25) 20	3.0	2.9	2.8	1.7	9.9	3.9	4.4
Barium	1	(1000) 750	766	763	772	609	903	834	854
Beryllium	0.02	1.2	1.40	1.40	1.38	1.00	1.77	1.47	1.54
Bismuth	0.02	–	0.08	0.06	0.10	0.01	0.20	0.10	0.10
Cadmium	0.01	12	0.30	0.27	0.27	0.11	0.75	0.44	0.59
Calcium	5	–	26978	26396	25550	19300	54300	30800	37100
Chromium	0.5	(1000) 750	44.8	44.0	43.4	28.8	74.5	56.7	58.6
Cobalt	0.10	(50) 40	8.36	8.18	8.05	5.55	15.18	10.05	11.58
Copper	0.50	(300) 225	13.19	12.28	12.10	6.27	42.46	18.63	19.42
Iron	20	–	21481	21079	20750	15300	33200	25950	28110
Lead	0.05	200	64.69	42.29	33.78	15.60	547.44	107.23	205.40
Lithium	0.1	–	11.3	10.9	10.5	7.4	21.7	16.2	18.5
Magnesium	1	–	8937	8816	8890	6240	12000	11010	11529
Manganese	0.2	–	525.3	512.1	531.6	320.4	872.9	662.4	718.1
Mercury	0.005	10	0.107	0.055	0.048	0.018	2.013	0.089	0.111
Molybdenum	0.10	40	0.64	0.59	0.60	0.30	1.70	0.91	1.26
Nickel	0.2	(200) 150	16.3	15.9	15.8	10.5	27.9	22.0	23.1
Phosphorus	10	–	1240	1172	1160	570	2710	1723	1909
Potassium	20	–	18035	17983	18150	13400	20900	19320	20588
Rubidium	0.1	–	52.0	51.0	48.4	36.3	91.1	65.0	78.2
Selenium	0.5	10	0.7	0.6	0.7	0.3	1.2	0.9	0.9
Silver	0.01	(25) 20	0.31	0.28	0.30	0.20	1.50	0.40	0.43
Sodium	10	–	22042	21929	22200	16500	26200	25150	25710
Strontium	0.1	–	360	359	356	288	437	401	418
Tellurium	0.02	–	0.04	0.02	0.01	0.01	0.20	0.10	0.10
Thallium	0.01	4.1	0.29	0.29	0.32	0.21	0.42	0.33	0.36
Tin	0.10	–	1.65	1.41	1.32	0.77	11.11	1.98	2.65
Uranium	0.01	–	1.17	1.11	1.10	0.66	2.64	1.56	1.96
Vanadium	0.5	(250) 200	46.8	44.9	45.7	28.5	85.6	68.1	70.6
Zinc	1.0	(800) 600	113.7	101.6	100.2	50.4	380.4	179.7	222.6

<sup>a</sup>All elements were determined by ICP-MS except mercury, which was determined by cold vapor AAS.<sup>b</sup>MDL = minimum detection limit; concentrations were above MDL for all elements except bismuth in 10 samples and tellurium in 31 samples; samples below MDL were assigned a value = 0.5 × MDL.<sup>c</sup>Soil Quality Criteria for residential/parkland from OMEE (1996). Criterion values in brackets apply to medium and fine textured soils.



Table 3  
Total element concentrations (mg/kg) in housedust of Ottawa residences ( $n = 48$ ).

Element <sup>a</sup>	MDL <sup>b</sup> (mg/kg)	No. samples < MDL	Arithmetic mean	Geometric mean	Median	Minimum value	Maximum value	90th percentile	95th Percentile
Aluminum	5	None	25948	24281	22900	12700	51100	40160	44225
Antimony	0.02	None	7.28	5.54	5.12	1.16	57.41	12.50	15.38
Arsenic	0.5	None	7.3	4.9	4.1	1.7	79.5	12.8	18.5
Barium	1	None	492	454	442	190	1480	682	803
Beryllium	0.02	None	0.56	0.53	0.53	0.28	1.00	0.81	0.90
Bismuth	0.02	None	1.67	1.02	0.79	0.21	8.62	3.78	6.48
Cadmium	0.01	None	6.46	4.42	4.30	1.12	34.94	15.30	17.32
Calcium	5	None	48760	46714	45250	29400	117200	65300	66770
Chromium	0.5	None	86.7	75.4	69.2	33.5	330.3	157.3	191.8
Cobalt	0.10	None	8.92	8.40	8.77	3.28	22.67	12.05	13.10
Copper	0.50	None	206.08	170.69	157.30	59.36	601.47	381.76	488.96
Iron	20	None	14135	13156	13150	7060	40350	19120	21575
Lead	0.05	None	405.56	232.61	222.22	50.20	3225.66	969.37	1311.92
Lithium	0.1	None	6.3	6.1	6.1	3.7	15.5	7.9	8.2
Magnesium	1	None	9826	9442	9285	5130	23250	12895	13390
Manganese	0.2	None	269.3	260.3	266.5	136.9	423.5	365.8	407.3
Mercury	0.005	None	3.633	1.728	1.607	0.012	37.099	6.565	12.558
Molybdenum	0.10	None	3.16	1.96	1.70	0.66	28.64	3.78	14.22
Nickel	0.2	None	62.9	53.6	51.5	16.0	243.3	103.4	116.4
Phosphorus	10	None	1380	1335	1268	890	3600	1761	1936
Potassium	20	None	10305	10021	10060	5715	15900	13530	14575
Rubidium	0.1	None	25.3	24.6	24.8	15.6	40.2	34.6	34.9
Selenium	0.5	5	1.2	1.0	0.8	0.3	6.8	2.0	2.2
Silver	0.01	None	2.05	1.48	1.30	0.40	9.33	4.46	6.50
Sodium	10	None	23224	20359	17970	10705	70580	46629	57380
Strontium	0.1	None	255	242	249	130	410	369	382
Tellurium	0.02	1	0.08	0.07	0.07	0.02	0.28	0.12	0.13
Thallium	0.01	None	0.14	0.14	0.13	0.08	0.24	0.20	0.21
Tin	0.10	None	54.84	21.87	16.87	2.66	595.02	111.12	221.33
Uranium	0.01	None	0.58	0.55	0.54	0.29	1.33	0.85	1.06
Vanadium	0.5	None	24.8	23.7	22.0	13.1	43.6	35.5	39.9
Zinc	1.0	None	716.9	628.0	633.1	239.1	1840.0	1226.0	1460.8

<sup>a</sup>All elements were determined by ICP-MS except mercury which was determined by cold vapor AAS.

<sup>b</sup>MDL = minimum detection limit; samples below MDL were assigned a value =  $0.5 \times \text{MDL}$ .

### 3.3. Differences amongst exterior and indoor media

In Ottawa, the multi-element signature of indoor dust differs significantly from that of garden soil and street dust (Fig. 2). The histograms in Fig. 2 were derived by normalizing the 90th percentiles of each of the three urban dust data sets reported in this study (Tables 2–4) to the 90th percentiles of natural background concentrations determined in the clay size-fraction of soil parent-material of the surrounding region (Kettles and Shilts, 1994). Although the histogram in Fig. 2 includes only the 14 elements that were

reported by Kettles and Shilts (1994), the available data highlight the pronounced differences in metal concentrations between indoor dust and exterior dust and soils. The histogram in Fig. 2 also places the urban data into the context of natural background concentrations in south-eastern Ontario, indicating that, for many elements, housedust concentrations exceed natural concentrations. These comparisons may be considered semi-quantitative owing to methodological differences between the two studies, the geological data being derived from a partial digestion of a finer particle size fraction. For lead, cadmium,

Table 4  
Total element concentrations (mg/kg) in street dust adjacent to Ottawa residences ( $n = 45$ )

Element <sup>a</sup>	MDL <sup>b</sup> (mg/kg)	No. samples < MDL	Arithmetic Mean	Geometric Mean	Median	Minimum value	Maximum Value	90th percentile	95th percentile
Aluminum	5	None	47540	46462	47700	12200	62300	57020	58060
Antimony	0.02	None	0.89	0.44	0.42	0.09	15.88	1.12	1.62
Arsenic	0.5	8	1.3	1.0	1.4	0.0	2.9	2.4	2.5
Barium	1	None	576	564	584	153	714	684	687
Beryllium	0.02	None	0.97	0.95	1.00	0.31	1.25	1.09	1.12
Bismuth	0.02	4	0.05	0.04	0.04	0.01	0.22	0.06	0.11
Cadmium	0.01	None	0.37	0.33	0.30	0.08	1.12	0.63	0.79
Calcium	5	None	96787	92654	95100	18200	198200	121420	132100
Chromium	0.5	None	43.3	41.7	41.8	14.7	71.7	58.5	63.9
Cobalt	0.10	None	8.31	8.02	8.65	2.31	12.59	10.80	11.15
Copper	0.50	None	65.84	38.13	29.54	4.79	249.78	187.71	236.18
Iron	20	None	18948	18210	18000	7300	33400	25660	27700
Lead	0.05	None	39.05	33.49	32.93	12.63	122.35	68.35	85.30
Lithium	0.1	None	7.4	7.3	7.6	2.4	10.1	8.5	8.7
Magnesium	1	None	15780	15086	15100	5800	27900	21460	23010
Manganese	0.2	None	431.5	420.0	426.4	145.1	618.3	533.7	581.5
Mercury	0.005	1	0.029	0.019	0.018	0.004	0.188	0.060	0.096
Molybdenum	0.10	None	1.39	1.29	1.38	0.38	2.68	1.98	2.16
Nickel	0.2	None	15.2	14.8	14.6	4.7	25.0	19.0	19.4
Phosphorus	10	None	616	597	606	344	951	840	875
Potassium	20	None	14963	14709	15000	4000	18300	17180	17280
Rubidium	0.1	None	37.2	36.3	37.8	9.8	45.4	42.9	44.5
Selenium	0.5	24	0.5	0.4	0.5	0.1	1.1	0.7	0.8
Silver	0.01	None	0.22	0.18	0.16	0.07	2.18	0.24	0.30
Sodium	10	None	18273	17220	17000	4200	54800	23220	27540
Strontium	0.1	None	459	446	445	92	735	539	567
Tellurium	0.02	1	0.08	0.07	0.08	0.01	0.25	0.13	0.14
Thallium	0.01	1	0.21	0.19	0.21	0.01	0.32	0.24	0.25
Tin	0.10	None	3.02	1.64	1.19	0.30	25.09	8.38	10.30
Uranium	0.01	None	0.82	0.79	0.80	0.43	2.25	0.97	1.02
Vanadium	0.5	None	34.0	32.2	34.2	13.8	56.2	48.3	50.6
Zinc	1.0	None	112.5	101.3	98.7	28.7	302.5	184.2	194.2

<sup>a</sup>All elements were determined by ICP-MS except mercury, which was determined by cold vapor AAS.

<sup>b</sup>MDL = minimum detection limit; samples below MDL were assigned a value =  $0.5 \times \text{MDL}$ .

mercury and silver, the 90th percentiles of house-dust concentrations are approximately an order of magnitude higher than the 90th percentile of natural concentrations. Concentrations of nickel, chromium, arsenic, copper and zinc in housedust exceed the natural concentrations by a smaller margin, with factors ranging between 1.2 and 5.6 (Fig. 2). In the case of molybdenum, cobalt, iron, manganese, and uranium, however, the pattern is reversed, in that 90th percentile natural concentrations exceed 90th percentile housedust concentrations by a factor of 1.5–6.7 (Fig. 2).

In contrast to housedust, concentrations in garden soil and street dust are low in comparison to local natural concentrations for most of the elements shown in Fig. 2. In garden soil samples, natural concentrations are exceeded only in the case of lead, cadmium, and silver (Fig. 2). For garden soil samples, 32 out of 50 samples (64%) exceed the 90th percentile natural lead concentration (28 mg/kg). Three residences have garden soil lead concentrations that range from 225–547 mg/kg, thus exceeding the Ontario Ministry of Environment and Energy (OMEE, 1996) residen-

tial soil criterion for lead (200 mg/kg). All three are located in older neighborhoods. No garden soil samples exceed the Canadian Council of Ministers of the Environment recommended residential soil guidelines of 10 mg/kg for cadmium (CCME 1997, 1999) or 20 mg/kg for silver (CCME 1991). With respect to street dust, 90th percentile natural concentrations are exceeded for only two elements in Fig. 2 (lead and cadmium). For the remainder of the elements in Fig. 2, higher concentrations are encountered in natural media (soil parent material) of the surrounding region than in street dust or garden soil of the city of Ottawa.

### 3.4. Variations according to house age and style of heating

The data show that, in general, higher lead concentrations occur in dust samples of older homes. The relationship between housedust lead content and house age has been observed elsewhere (Crocetti et al., 1990; Thornton et al., 1994; Sutton et al., 1995; CMHC, 1997; Meyer et al., 1999), and is attributed primarily to a decrease in the use of lead-based paint, solders, and plumbing materials since the Second World War (CMHC,

1997). For the subset of 25 gas-heated homes built in the 1900s, linear regression analysis yields a correlation of  $R^2 = 0.61$  (95% confidence level) between the concentration of lead in housedust and the age of the house. There was a lack of correlation between housedust lead content and house age in the subset of electrically heated homes ( $R^2 = 0.01$ ;  $n = 7$ ) and the subset of oil-heated homes ( $R^2 = 0.17$ ;  $n = 14$ ). The observation that this correlation holds only for gas-heated houses, and not for the whole data set ( $R^2 = 0.20$ ;  $n = 48$ ) does suggest that factors other than age can have an overriding influence.

A comparison of housedust metal concentration data divided according to style of heating (Table 5) shows that housedust of electrically-heated houses tends to be higher in both lead and mercury content than housedust of gas- or oil-heated houses. These results suggest that the method of heating the house has an effect on metal accumulation in housedust, possibly due to the associated differences in air circulation and use of particle filters. Thus, it is not necessarily true that lower concentrations of all metals can be expected in housedust of newer homes. The median mercury content of housedust in post-1950

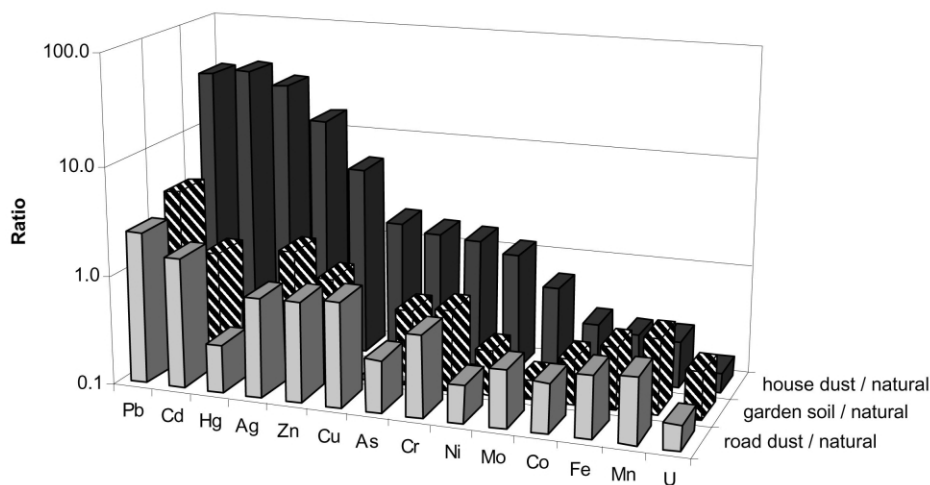


Fig. 2. Multi-element signatures of housedust, street dust and garden soils, derived by normalizing the garden soil, housedust, and street dust concentrations (Tables 2–4) to natural background concentrations for soil parent material of the same region (Kettles and Shilts, 1994). Calculations used 90th percentiles of all four data sets. Background concentrations were determined using aqua regia digestion of the  $< 2\text{-}\mu\text{m}$  fraction of parent material collected 1 m below surface at  $> 1700$  sites in south-eastern Ontario and south-western Quebec.

Table 5  
Concentrations of lead and mercury in housedust of Ottawa residences according to age and style of heating<sup>a</sup>

	Concentration in housedust (mg/kg)				
	Age		Type of heating		
	Pre-1950	Post-1950	Electric	Gas	Oil
<b>Lead (mg/kg)</b>					
<i>N</i>	19	29	7	27	14
Mean	619	266	462	389	409
S.D.	619	331	418	623	508
Median	342	135	344	222	198
<b>Mercury (mg/kg)</b>					
<i>N</i>	19	29	7	27	14
Mean	3.35	3.82	8.97	3.01	2.26
S.D.	3.35	6.69	12.92	4.48	12.40
Median	1.27	2.79	4.13	1.36	1.39

<sup>a</sup>At the time of survey (1993) median house age was 41 years for gas-heated, 40 years for oil-heated, and 18 years for electrically heated houses.

homes, for example, is more than double that of homes built in 1950 and earlier (Table 5). As indicated earlier, the median age of electrically heated houses was 18 years at the time of the survey (1993), which is less than half the median age of houses heated by gas or oil.

#### 4. Discussion

In the city of Ottawa, housedust samples contain significantly higher concentrations of certain key elements, such as lead, cadmium, mercury and antimony, than either garden soil or street dust. This observation has important implications for source apportionment modeling, as it differs from other studies, which indicate that concentrations of these elements in housedust are in the same order of magnitude as their concentrations in soil (Fergusson and Kim, 1991). Although the results of this study do not permit us to draw any firm conclusions on the indoor sources of contamination, they do indicate that dust generated within the house itself can be an important source of exposure for certain elements. It is important to note that indoor/outdoor concentration ratios vary widely from one element to another, and

from one residence to another within the community. These variations, combined with the distinct multi-element signature of housedust compared to exterior soil and dusts in Ottawa (Fig. 2), make it difficult to accurately predict the contribution that soil makes to element concentrations in housedust.

The US EPA Integrated Exposure Uptake Biokinetic (IEUBK) Model for Lead in Children recommends measurements of indoor lead concentrations for residential assessments, but in the absence of household dust data the model assumes that soil is the predominant source of lead in indoor dust (US EPA, 1998). To approximate the concentration of lead in indoor dust the *Multiple Source Analysis* module of the IEUBK model provides a default estimate of 0.70 g soil/g dust, expressed as a ratio of indoor dust lead concentration to outdoor soil lead concentration (US EPA, 1998). The results of the present study indicate that this default estimate is not applicable to the city of Ottawa. In this case, lead concentrations in housedust are approximately five-fold higher than lead concentrations in garden soil (using either geometric means or medians in the calculation), and there is a lack of correlation between lead concentrations in indoor dust and outdoor soil ( $R^2 = 0.065$ ) due to the variability of indoor/outdoor ratios. The median concentration ratio for lead in housedust/lead in garden soil is 5.4 for the five post-1980 residences, which is approximately the same as the median ratio for houses built before 1980 (median ratio of 5.0 for 43 pre-1980 houses). Given the relatively low range of lead concentrations in Ottawa soils observed in this study and in the previous study of Ericson and Mishra (1990), application of the default estimate (0.70 g soil/g housedust) would result in a significant underestimation of indoor dust lead concentrations in comparison to the actual indoor concentration data reported here. The potential for error in this case underscores the importance of collecting separate indoor versus outdoor environmental quality data for residential exposure assessments.

Previous studies have indicated that the style of heating influences indoor-dust metal concentrations and loading rates, with higher metal concen-

trations attributed to the use of coal and other fossil fuels as the main source of heat (Meyer et al., 1999). The style of heating was also found to influence indoor-dust metal concentrations in this survey (Table 5), but there is a difference, in that coal is not used, and concentrations of lead and mercury are actually higher in dust samples from electrically heated houses than in dust samples from houses heated with other fossil fuels (oil and gas). This observation may be explained by lower rates of exchange between indoor and outdoor air in houses with passive baseboard or radiator electrical systems, compared to houses with oil and gas systems which use forced air and are fitted with particle traps.

Total element concentrations in dust are likely to be influenced by the relative proportion of biogenic particles (molds, fungi and other organic matter), which have an ability to accumulate metals to high concentrations. Fergusson and Schroeder (1985) reported much higher concentrations of organic matter in indoor dust (approx. 40%) compared to garden soil and street dust (approx. 9%). Inadequate exchange between indoor air and outdoor air can result in increased indoor fungal concentrations (Ren and Leaderer, 1999), and since fungi and other lower plants are capable of accumulating high concentrations of mercury (Rasmussen, 1994), it follows that higher

mercury concentrations may accumulate in the indoor dust of damp, poorly ventilated homes. In general, high indoor/outdoor concentration ratios of metals that have an affinity for organic matter may be partly attributable to the higher organic content of housedust compared to garden soil and street dust.

In a previous estimate of total daily lead exposure for a 2-year-old (13.6 kg) Canadian child, Health and Welfare Canada (HWC, 1992) concluded that soils and housedust are significant sources of exposure (Fig. 3a). In their estimate, dust and dirt together account for 27% of total daily lead uptake, based on an ingestion rate of 80 mg of dirt/day at a concentration of 140 mg/kg lead, adjusted by an absorption factor of 30% (HWC, 1992). Recently, White et al. (1998) recommended disaggregating the allocation for dirt ingestion into separate categories for exterior dirt and indoor housedust. The effect of implementing this recommendation in the present study is shown in Fig. 3b,c, wherein the single dirt ingestion portion of the original estimate (Fig. 3a) is disaggregated by substituting lead concentration data from Tables 2 and 3 and allocating 71% (57.1 mg) of total dirt ingestion to indoor dust and 29% (22.9 mg) to exterior soil. Fig. 3b is derived using the geometric mean lead concentration of garden soil (42.3 mg/kg; Table 2) for the

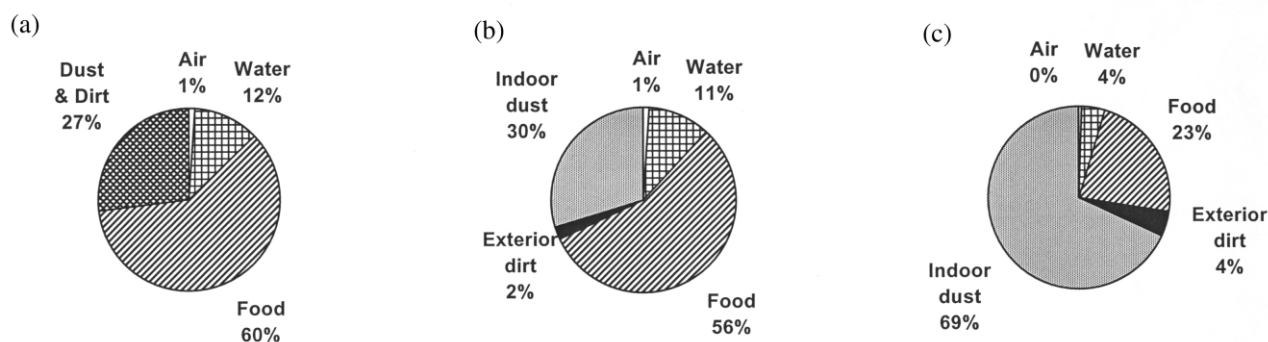


Fig. 3. The effect of disaggregating lead exposure from dirt ingestion into separate indoor dust and exterior soil allocations. Pie diagram (A) represents the exposure apportionment for a 2-year old (13.6 kg) child from HWC (1992). The dust and dirt allocation assumes an intake of 80 mg dirt/day and 30% absorption. Pie diagram (B) is the same as (A) except dirt ingestion is separated into exterior soil and indoor dust components, using geometric mean lead concentrations from Tables 2 and 3. Pie diagram (C) is the same as (B) except that 95th percentile lead concentrations from Tables 2 and 3 were substituted for the exterior soil and indoor dust components.

exterior dirt portion, and the geometric mean lead concentration for housedust (232.6 mg/kg; Table 3) for the indoor dust portion. While this disaggregation does not substantially alter the combined contribution of dirt and dust ingestion from the original estimate (32% of total daily uptake; Fig. 3b), it does reveal housedust to be the second largest contributor to total daily lead uptake after food (30 and 56%, respectively; Fig. 3b). In Fig. 3c, the 95th percentile lead concentrations from Tables 2 and 3 are used, instead of the geometric mean lead concentrations, to represent the most likely maximum concentrations that would be encountered in and around an Ottawa residence. In this case, the allotment for indoor dust ingestion increases to 69% of the total (Fig. 3c), representing the largest single contributor to a child's daily lead exposure.

More precise estimates of children's exposures may be obtained through the use of hand wipes or indoor dust loading measurements (Duggan and Inskip, 1985; Gulson et al., 1995; Lanphear et al., 1998). The primary goal of this study was to compare the multi-element profile of indoor dusts to that of exterior dusts and soils, and the sample collection and preparation protocols described herein were designed to meet that goal. However, if the total concentration data in Tables 2–4 are to be used in the context of exposure assessments and the development of indoor dust guidelines, appropriate absorption factors (coefficients to approximate bioavailability) should be applied. In Fig. 3, for example, an absorption factor of 30% was applied in the calculation of the dust and soil allocation. As indicated earlier, hydrofluoric acid was used for the digestion of all three sample media and, additionally, perchloric acid in the case of street dust. These are strong reagents, which are not representative of either salivary or digestive fluids, and therefore an assumption of 100% bioavailability would result in an overestimate of gastrointestinal uptake. Absorption factors vary widely, depending on the element or compound under assessment, the particle size, the age of the receptor, and the time elapsed since the last meal. In the next phase of this work, sequential extraction techniques will be applied to different particle size fractions of Ottawa dust

and soil samples to allow better estimates of the amount of each element that may be available for human absorption.

## 5. Conclusions

Results of the multi-element profiles of indoor dust versus exterior soils and dusts collected from 50 residences located in 10 neighborhoods across the city of Ottawa, capital city of Canada, indicate that household dust has a distinct multi-element signature compared to exterior soils and dusts, and from this it is concluded that dust generated within the house itself is an important source of exposure for certain elements. In addition to indoor sources of metals, the proportion of biogenic particles such as fungi and molds, which have a high affinity for metals, may be an important factor influencing the distribution of element concentrations in indoor dust. Because of the wide variations in indoor/outdoor concentration ratios from one element to another in the same residence, and from one residence to another within the community, it would be extremely difficult to accurately predict indoor dust concentrations based on exterior soil data.

The present results underscore: (i) the importance of obtaining separate measurements for indoor dust and exterior dirt to improve residential exposure assessments; and (ii) the validity of developing a separate set of guidelines for elemental concentrations in indoor dust. In 1991 the Canadian Council of Ministers of the Environment (CCME) lowered the recommended guideline for lead in residential soil to 500 mg/kg (from 1000 mg/kg), and in 1997 lowered it further to 140 mg/kg, in an effort to address the health implications of exposure of young children to environmental lead (CCME, 1997, 1999). However, the present study suggests that only a small proportion (2–4%) of the total daily lead exposure arises from ingestion of exterior soil, while a far more significant portion may be attributed to ingestion of household dust. Our calculations indicate that indoor sources could account for at least 30% of total daily exposure if geometric means are used (Fig. 3b) or 69% if 95th per-

centiles are used (Fig. 3c). Therefore, for a city such as Ottawa with few industrial sources, a significant reduction in childhood exposure to lead and other elements of concern, such as mercury, cadmium and antimony, will not be accomplished through continued lowering of exterior soil clean-up criteria and guidelines, but through increased attention to indoor sources of exposure, and improved parental attention to personal hygiene and housekeeping practices.

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