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Wipe Sampling Methodologies to Assess Exposures to Metals in Urban Canadian Homes

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Wipe Sampling Methodologies to Assess Exposures to Metals in Urban Canadian Homes

1. Cadmium, Lead, Yttrium

2. Antimony, Arsenic, Chromium, Copper, Nickel

By: Lauren McDonald

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Abstract

Wipe sampling is a means of monitoring exposures to contaminants which has been used extensively for assessment of lead (Pb) in housedust in the U.S. This thesis reports the first baseline wipe dataset for urban Canadian homes, for Pb and other metal(loid)s including: arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), and antimony (Sb). The literature review covers sampling methods, health effects, and sources. The digestion procedure ASTM 1644 is the accepted HNO_3 hotplate protocol for determination of Pb in wipes. However in this study, HF was added to enhance extraction efficiencies for a wide range of elements. The first paper identifies baseline and elevated subpopulations for Cd and Pb using normality plots, and investigates the use of yttrium(Y) as a soil tracer. The second paper investigates application of the wipe methodology for monitoring As, Cr, Cu, Ni, Sb, and Zn. This is the first multi-element wipe sampling dataset in the literature, and will assist risk assessors and risk managers in providing guidance to Canadians about ways to reduce exposures to metals.

Résumé

L'échantillonnage avec lingettes est une technique employée afin d'étudier l'exposition aux contaminants et est largement utilisée aux États-Unis pour la détermination de l'exposition au plomb (Pb) contenu dans la poussière domestique. Cette thèse rapporte la première série de données obtenues selon un échantillonnage avec lingettes pour le niveau de base en plomb et autres métaux tel que l'arsenic (As), le cadmium (Cd), le chrome (Cr), le cuivre (Cu), le nickel (Ni) et l'antimoine (Sb) dans les maisons urbaines au Canada. La révision de la littérature comprend: les méthodes d'échantillonnage, les effets sur la santé et les sources. La méthode de digestion ASTM 1644 est le protocole sélectionné pour la détermination du taux de plomb dans les lingettes. Cette méthode utilise une plaque chauffante et de l'acide nitrique (HNO_3). Pour les fins de cette étude, de l'acide fluorhydrique (HF) a été ajoutée afin d'améliorer le rendement de l'extraction pour une variété de métaux. Le premier article indique le niveau de base et les sous-populations présentant des valeurs élevées pour le Cd et le Pb en utilisant les graphiques de normalité et discute de l'utilisation de l'yttrium comme élément traceur pour le sol. Le second article examine l'utilisation de la méthode des lingettes pour la surveillance de l'exposition aux éléments suivants : As, Cr, Cu, Ni, Sb et Zn. Ceci représente la première série de données pour l'échantillonnage avec lingette publiée dans la littérature. Ces informations aideront les évaluateurs et les gestionnaires du risque pour guider les Canadiens dans les méthodes de réduction de l'exposition aux métaux.

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List of Publications

This is a paper-format thesis. It is based on work presented in the following publications:

I Lauren T. McDonald, Pat E. Rasmussen, Marc Chénier, Christine Levesque
Wipe Sampling Methodologies to Assess Exposures to Lead and Cadmium in Urban Canadian Homes
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II Lauren T. McDonald, Pat E. Rasmussen, Marc Chénier, Christine Levesque
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Published Abstracts and Presentations

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Canadian Environment and Child Health and Development Research Workshop
February 9th, 2009 "Childhood Exposures to Metals in Urban Canadian Shield Homes Assessed Using Wipe Sampling Methodologies"
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Earth and Health Collaboration Launch Event, University of Ottawa
June 10th, 2009. "Exposures to Metals in Urban Canadian Shield Homes Assessed Using Wipe Sampling Methodologies"
University of Ottawa, Ottawa, ON. Poster Presentation.

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269 Laurier Ave, Ottawa, ON. Health Canada. Oral Presentation.

Health Canada Science Forum
November 16th – 17th, 2009. "Wipe Sampling Methodologies to Assess Exposures to Lead and Cadmium in Urban Canadian Homes"
Hampton Inn, Ottawa, ON. Abstract and Poster Presentation.

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Literature Review

Introduction

Health Canada (2009) estimates that the average Canadian spends 90% of their time indoors. Therefore it is important to be aware of the contaminants in the built environment and the routes of exposure to them. This research contributes to the understanding of exposures to metals mainly through the ingestion of house dust. This is particularly of concern with respect to small children who crawl on the floor. Their exploratory nature results in normal hand-to-mouth behaviour, ingesting an estimate of 100 milligrams of dust per day (Lewis et al. 1994). The USEPA (2008) has published a lower estimate of dust ingestion; the value is 30 mg/day for infants 6-12 months and 60 mg/day for children one year of age up to 21 years. The existing literature on wipe sampling focuses mainly on lead (Pb) however there are many other metals that pose health risks as well. McDonald et al. (2009a, under review) examines exposures to cadmium (Cd) as well as Pb, while McDonald et al. (2009b, under review) focuses on arsenic (As), chromium (Cr), copper (Cu), nickel (Ni), and antimony (Sb).

Composition of House Dust

Dust is defined as a heterogeneous matrix of particles varying in size that accumulates in the home; its composition varies from home to home and room to room (Park et al. 2000). Some common components of residential house dust include: asbestos, bacteria, dead skin, dust mites and their fecal matter, fibres from clothing and furnishings, food, human and animal hair, insects, minerals, mould spores, metals (Cd, Cr, Cu, Ni, Pb etc), paint chips, paper, pesticides, pet dander, plastics, pollen, rubber, sand, etc (Molhave et al. 2000; Yu et al. 2006; Turner and

Simmonds 2006; Turner and Ip 2007). A study of seven Danish office buildings identified several minerals in vacuum dust by scanning electron microscope which included: apatite, biotite, calcite, feldspar, gypsum, ilmenite, halite, hematite, hornblende, quartz, limonite, and pyrite (Molhave et al. 2000). Minerals occur primarily in the coarse fraction (80-300 μm) of house dust (Woldemichael et al. 2009). House dust from homes in the three cities examined in the current study was also analysed for minerals. Similar mineralogy was observed despite differences in bedrock geology. It was the surficial geology that was the dominant factor, contributing quartz (from de-icing in winter months) to house dust (Woldemichael et al. 2009). Metals are however the focus of the current research and they occur most abundantly in the finest fraction (< 80 μm) of house dust (Rasmussen 2004a).

Exposure Assessment

Toddlers commonly crawl on the ground and put their hands in their mouths. Thus they have an increased risk of exposure to the contaminants in house dust. Therefore researchers have aimed to quantify this exposure (Rodes et al. 2001). It has been suggested that particles less than 100 - 200 micrometers in diameter are those that children's hands typically acquire (Adgate et al. 1998; Sheppard and Evenden 1994). Thus metals are prime candidates to adhere to a child's hand during normal daily activities. Metals, in minute quantities, are often necessary for optimal growth and health; however overdoses can pose dreadful health consequences. Infants and children are most susceptible due to their early brain development (Tong and Lam 2000).

Pica refers to the excessive consumption of non-dietary substances and is most common in children aged six to twelve (Roberts et al. 1992). This is a medical condition which occurs in up to 30% of children at some point in their childhood (Gavin 2007). Such substances may include but are not limited to: dirt, dust, feces, glue, paint chips, paper, and soap (Gavin 2007).

More specifically, geophagia is the custom where a person deliberately ingests earth, soil, and or clay (Woywodt and Kiss 2002). These conditions are of concern due to the ingestion of potentially harmful contaminants within these substances. Paint chips, for example can contain Pb, depending on the age of the paint, and when ingested, are a common cause of an elevated blood Pb level in a child. This of course is a problem even with non-pica children as paint chips break down and contribute to Pb in house dust. These two conditions (i.e. pica and geophagia) result in higher levels of metals in the body compared to the exposure from normal hand-to-mouth behaviour of unaffected children.

It has been estimated that the average person spends eighty to ninety-five percent of their time indoors (Binder et al. 1987; Health Canada 2009; Butte and Heinzow 2002). With these facts in mind, thorough cleaning of the home, specifically with respect to reducing dust levels, is critical especially when small children reside in a home. The removal of shoes upon entering the home is also essential (Roberts et al. 1999). Soil and street dust is estimated to contribute anywhere from 20 – 95 percent of house dust (Meyer et al. 1999a; Rasmussen et al. 2001). Soil and street dust consist of metals from various sources including land use and geological weathering (Al-Khashman 2007). Thus many contaminants enter the home from outdoors via various pathways.

Sampling Procedures

Wipes

Wipe sampling is an excellent means of sampling house dust for the purpose of exposure assessments. The technique of wipe sampling also referred to as swipes or smear sampling is a method of dust collection gaining in popularity. It has mainly been employed for sampling Pb in the United States (Adgate et al. 1998; Lanphear et al. 1998) with some work also done on Cr

(Lioy et al. 1992; Freeman et al. 1997). Several governing bodies have released protocols and/ or regulations with regard to wipe sampling procedures. These include the American Society for Testing and Materials (ASTM), the Department of Housing and Urban Development (HUD), the Environmental Protection Agency (EPA), the National Institute for Occupational Safety and Health (NIOSH), and the Occupational Safety and Health Association (OSHA). The HUD Pb clearance level for floors was initially set at 100 micrograms per square foot ($\mu g\ ft^{-1}$), measured by wipe sampling, reflecting their requirement for wipes as the sampling medium (USHUD 1995). However the EPA adjusted these levels in part due to work and criticism by Lanphear et al. (1998) and Sterling et al. (1999). The floor Pb dust regulation was subsequently reduced to 40 $\mu g\ ft^{-1}$ (USEPA 2000). This value however has also been criticized and recommendations have been made in the literature to have it lowered once again. Dixon et al. (2009) recommended 12 $\mu g\ ft^{-1}$ Pb in floor dust to protect the majority (95%) of children from having elevated blood Pb levels.

Many researchers are in support of the regulatory wipe method. For the purpose of determining individual exposures to residential sources of Pb, many researchers view wipe sampling as superior to vacuum sampling. Wipes provide the researcher with a loading value reported as mass of contaminant per unit surface area. Vacuum sampling alternatively yields a concentration value reported as mass of contaminant per mass house dust. Loadings (e.g. Pb loadings) have thus been identified to provide more relevant information on exposures (Lanphear 2008). Wipe sampling is also advantageous as it allows one to assess room-to-room variability. This then allows for the identification of sources based on the activities conducted in the various rooms sampled. For example, a wipe collected in the entry is more representative of sources entering the home from the outdoor environment. Likewise a sample collected from the second floor of a home such as from a child's bedroom is more indicative of interior sources of metals in addition to soil tracked in.

Wipe sampling mimics a child's hand contact with hard surfaces and it is thus argued that a child has greater potential to be exposed to Pb from smooth versus carpeted surfaces (Yiin et al. 2002). Caplan et al. (1993) stated that wipe samples are a good approximation of skin absorption. Rodes et al. (2001) studied the percentage of the hand that actually makes contact with a surface and found it to be merely 33 percent. Therefore, sampling with a vacuum may not be representative of tangible exposure compared to sampling with wipes, especially on non-carpeted surfaces.

The question of where to sample was studied by Wilson et al. (2006) as the EPA does not specify the best location(s) and number of sample(s) to be taken within the home, to most accurately represent exposure within it. Wilson et al. (2006) found the best predictor of elevated blood Pb in a child to be from non-carpeted floor wipe samples from the home's entry way, kitchen, living room, and bedrooms. Floor samples, from these locations, were found to be sufficient to determine Pb loadings for the whole home. This work found bathroom and windowsill sampling to be less strongly associated with a child`s blood Pb level (Wilson et al. 2006). Windowsills have been extensively sampled in the past, therefore this conclusion could have a significant impact on the field.

Health Effects of Study Elements

It is important to note that the elemental composition of house dust can vary depending on location based on many factors including geology, industrial activity, and wind patterns. The three study cities are Barrie, Greater Sudbury, and Thunder Bay, Ontario. The elements that were chosen for analysis in this study include: arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), antimony (Sb), yttrium (Y), and zinc (Zn). These elements however must first pass quality assurance and control criteria to be included. These elements were chosen for several reasons. Firstly, they are the elements that Health Canada's Contaminated Sites Program is concerned with. Secondly, they have the potential to cause adverse human health effects. Lastly, they were chosen based on anthropogenic activity and land use in the three cities.

Arsenic (As)

The metalloid As occurs in three different oxidation states: metallic, trivalent, and pentavalent. It is present in silver and gold arsenide minerals as well as in the sulphide minerals arsenopyrite, realgar, and orpiment (Klein and Hurlbut 1999). Arsenic is associated with Cu and platinum group elements and is thus of specific concern in the city of Greater Sudbury where these are actively mined (Ames and Farrow 2007). Arsenic tends to be emitted into the environment during Cu smelting processes. Industrially, As is produced for use in wood preservation, insecticides, pigments, and semiconductors (Agency for Toxic Substances and Disease Registry (ATSDR) 2007; Cullen 2008).

Arsenic is a known human carcinogen that can result in cancer of the lung, bladder, and skin (Keegan et al. 2002). Chronic exposure to this element has been correlated to decreased cardiovascular health, diabetes mellitus, and reproductive effects (Wilhelm et al. 2005). Inorganic As is more toxic than organic As, which is commonly ingested via seafood and tobacco (Hysong et al. 2003). Levels of As in the body can be measured from first morning urine samples (Binder et al. 1987), or by finger nail clippings (Wilhelm et al. 2005).

In a study conducted by Benin et al. (1999) dust As concentrations were found to decrease with increasing distance from industrial activity. This was later confirmed by Keegan et al. (2002) who found that As levels dramatically decreased within five kilometres of Cu smelting activity. This trend is supported by the work of Hysong et al. (2003) in Arizona where they studied a Cu smelting town in relation to a town with no such industrial activity. They found that homes closer to the smelter had significantly higher concentrations of As in house dust than homes further away. This information pertains to this current research as the neighbourhoods sampled in Sudbury are located in varying distances to the mining sectors. Rieuwerts et al. (2006) stated that elevated As in the body is a result of ingestion of contaminated water and seafood, as well as ingestion, inhalation, and dermal absorption of soil and dust. Residents of Arizona, where Cu smelting occurs, were identified as being exposed to an average level of 69.2 micrograms per day of As whereas the ATSDR suggests the general public is typically exposed to 50 micrograms per day (O'Rourke et al. 1999). Arsenic has been identified as accumulating in house dust, as higher concentrations of this metalloid have been found in indoor dust versus garden soil or street dust (Lemus et al. 1996; Rasmussen et al. 2001). Sources of As in the home were investigated by O'Connor and Duncan (2008) and were determined to be minimal. They concluded that the majority of As enters the home from outdoor sources. However, the industrial sources of As listed above could be potential indoor sources.

Cadmium (Cd)

Cadmium occurs in two different oxidation states: metallic and divalent. It is typically present in association with Zn and forms two common sulphides: greenockite and hawleyite (Bernard and Hyrsl 2006). Due to its non corrosive properties, Cd is used in batteries, pigments, metal coatings, and plastics (Shoeters et al. 2006). It is more volatile than other heavy metals and is released during Pb and Zn processing (Bell 1998). The principle sources of Cd in the human body originate from food and tobacco (Yapici et al. 2006). The body absorbs three to seven percent but this number increases when iron levels are low, for example in pregnant women (Shoeters et al. 2006). Cadmium is a carcinogen specifically of concern to the lungs and thus of greatest toxicological concern when inhaled (Bussières et al. 2004). Cadmium is also nephrotoxic (Rasmussen and Gardner 2008a) and overexposure to this metal is highly toxic not only to the kidneys but it also disturbs calcium concentrations and reduces fertility (Shoeters et al. 2006).

Cadmium has been found to accumulate in house dust versus garden soil and street dust by several researchers around the world (Kim and Fergusson 1993; Lemus et al. 1996; Rasmussen et al. 2001; Al-Momani 2007). The enrichment was calculated to be 16 times greater in indoor dust versus outdoor soil in both: Ottawa, Canada, and Christchurch, New Zealand (Rasmussen 2004a). Sources contributing to and factors affecting Cd concentrations in house dust were summarized by Fergusson and Kim (1991) and include: red, orange, and yellow carpets, distance of home from road and industrial activity, dustiness and carpet wear, presence of a fireplace in the home, presence of a galvanized iron roof on the home, house age, preschool children living in the house, and rubber carpet underlay. Interestingly, Willers et al. (1993) found that tobacco and socioeconomic status were not relevant sources of Cd concentrations in house dust. Cadmium was found to be reduced in the home when floors were swept daily in a study in Hong Kong (Tong and Lam 2000).

Chromium (Cr)

From a geochemical perspective, Cr tends to substitute for trivalent aluminum and iron, thus it occurs in many silicates. Minerals containing Cr include: chromite and stichite (Bernard and Hyrsl 2006). Commercially, the metal is used in video and audio recording tapes, copy machine toner, dyes, paints, inks, and plastics (Lenntech 2008). Chromium is most toxic in its hexavalent form, and exposure can result in nosebleeds, liver and kidney damage, and lung cancer (OSHA 2008, ATSDR 2001). The EPA identifies Cr VI as a Group A, known human carcinogen via inhalation, OSHA sets the exposure limit at 0.05 milligrams per cubic meter (2008). The hexavalent form of Cr readily diffuses across the epidermis (National Research Council (NRC) 1976). Overexposure to Cr has also been found to weaken the immune system, cause ulcers and mutations, as well as respiratory problems.

Chromium has been found to accumulate in the indoor environment in house dust in relation to outdoor concentrations in soil and street dust in various geological settings (Lemus et al. 1996; Rasmussen et al. 2001; Al-Momani 2007). This is partly due to the fact that small Cr particles form aerosols, which are easily transported by wind up to several miles before settling (NRC 1976). In the past, the state of New Jersey, USA, has been an intense area of chromate production and thus exposure concerns have been investigated. A study by Lioy et al. conducted in 1992 found that 88% of homes surrounding industrial areas were contaminated with Cr versus only 25% in control homes located at least one half mile from industrial activity. They also found that wipes were better able to collect Cr from house dust than vacuuming due to its ability to collect all particles and characterize deposition on floor and upholstery (Lioy et al. 1992). A follow up study conducted five years later found that Cr could be reduced in homes by good housecleaning practices (Freeman et al. 1997).

Copper (Cu)

Copper is released to the environment via mining activities. Copper is being mined in Sudbury from a 60 by 30 kilometre elliptical depression likely formed from a meteor impact 1.85

billion years ago (Geological Survey of Canada 2008). Industrially, Cu is of great value as it is used in a wide variety of applications, including: cancer treatments, computers, currency, electrical wiring, fungicides, plumbing, and alloys of brass and bronze (ATSDR 2004). Food is the primary source of Cu to humans, and women with increased estrogen levels due to pregnancy or oral contraceptives possess higher levels of Cu (Bussières et al. 2004). Acute exposure to Cu via inhalation can result in nausea, vomiting, and irritation of the nose and throat (ATSDR 2004). Chronic overexposure can lead to brain, liver, and kidney damage, and ultimately death. It is however unknown whether Cu is carcinogenic (ATSDR 2004).

Copper has been found to accumulate in house dust versus outdoor soil by several researchers around the globe (Harper and Sullivan 1987; Kim and Fergusson 1993; Lemus et al. 1996; Rasmussen et al. 2001; Al-Momani 2007). In Christchurch New Zealand the indoor/outdoor ratio was calculated to be 10 while it was 14 in Ottawa, Canada (Rasmussen 2004a and references cited therein). Rasmussen et al. (2008b) found that Cu has a high affinity for organic carbon, this aids the explanation for the enrichment of Cu in house dust. Additionally, thirty-eight percent of Cu was determined to be in the form of Cu sulphide in the fine fraction of Ottawa house dust samples (Rasmussen et al. 2008b). Factors that have been found to increase Cu concentrations in house dust include: distance of home from road, dustiness, carpet wear, resident occupationally exposed, and high traffic density (Fergusson and Kim 1991). Enrichment in house dust is of concern due to its high bioaccessibility identified by Turner and Simmonds (2006), where Cu was found to be the most bioaccessible element followed by Pb, Zn, aluminum (Al), Ni, Cd, manganese (Mn), iron (Fe), and Sb.

Nickel (Ni)

Nickel is found in igneous rocks as its geochemistry allows it to substitute for divalent cobalt, iron, and magnesium. Breithauptite (NiSb), nickeline (NiAs) and pentlandite $(Fe,Ni)_9S_8$ are nickel minerals reported in Sudbury and Thunder Bay Ontario (Bernard and Hyrsl 2006). Pertinent to this study, Ni has been actively mined in Sudbury since 1885 and various companies have produced significant quantities of this metal. A recent human health risk assessment conducted by the Sudbury Soil Study (2008) revealed elevated air Ni levels in several regions of Greater Sudbury, quantifying levels to be as much as twice as high as the typical Ontario exposure. This is of concern as inhalation exposure to this heavy metal has been known to cause kidney, lung, nasal, and prostate cancers (Calabrese and Kenyon 1991). More commonly however, Ni causes allergic reaction when in contact with skin resulting in rashes (ATSDR 2005). Nickel has been found to accumulate to higher concentrations in house dust with respect to outdoor soil and street dust in Louisiana, U.S., and Ottawa, Canada (Lemus et al. 1996; Rasmussen et al. 2001). Bioaccessibility was studied by Rasmussen et al. (2008b) where Ni in indoor dust was found to be twice as bioaccessible as in outdoor soil.

Lead (Pb)

Lead averages 15 milligrams per kilogram in Earth's crust and it occurs in over 200 minerals however the most common are galena, anglesite, and cerussite (Bell 1998). It is ubiquitous in the urban environment and has severe health consequences thus it is the metal most thoroughly studied in the indoor environment. It has a long history of use by humans; however it was not until 1904 when Australian physician, J. Lockhart Gibson, noticed a connection between Pb in house paint and elevated blood Pb levels in children. He tested this observation by measuring the amount of Pb collected from wiping surfaces that were painted with paint containing high percentages of Pb. He found that ingestion was the primary source of exposure due to hand-to-mouth activity and that when the child was removed from the Pb painted home, blood Pb levels were reduced (Rosner et al. 2005). Unfortunately the high percentage of Pb in paint was not reduced in Canada until just several decades ago (CMHC 2009) despite this early discovery. Another source of exposure to Pb is via Pb based gasoline, although this was more quickly phased out. An estimate of 12 million metric tons of Pb was reported to be sold in the 20th century in the United States in the form of either a paint or gasoline additive (Mielke et al. 2008). Commercially, Pb is also used extensively in car batteries as well as in ammunition, and fishing equipment (ATSDR 2006).

Lead toxicity can be caused by inhalation and ingestion routes; the respiratory tract absorbs approximately 40 percent of inhaled Pb while the gastrointestinal tract absorbs merely 5 – 10 percent of ingested Pb (Harper et al. 2002). The Pb species dictates its bioaccessibilty, Pb carbonate for example is a form of Pb that is easily inhaled and absorbed in the gastrointestinal tract (van Alphen 1999). Lead poisoning has been meticulously studied due to the seriousness of the condition with effects on multiple systems of the body. Lead affects the nervous system by lowering intelligence quotients (Jusko et al. 2008) and it has been correlated with attention-deficit / hyperactivity disorder (Froehlich et al. 2009). Lead has also been correlated to violent behaviour such as murder and rape (Nevin 2000). It also affects the excretory, cardiovascular, and haematological systems possibly causing anaemia. In addition, Pb exposure affects the reproductive system causing reduced sperm count, infertility, decreased birth weight and miscarriages. Lead poisoning has been ultimately known to cause death (Harper et al. 2002; ATSDR 2006).

Childhood Pb exposure is currently of utmost concern in low income housing and also while redecorating and renovating older homes (Lanphear 2005). Children absorb Pb at five fold the rate of adults (Markel 2007). A pooled analysis conducted by Lanphear et al. (1998) found that children are susceptible to elevated blood Pb at floor dust Pb loadings of merely 5 – 10 μg ft^{-1}. The authors also identified that Pb-contaminated house dust was the source of low to medium elevated blood Pb levels in children. A child's blood Pb peaks at the age of two, however by this time their hand-to-mouth behaviour typically decreases. Intervention occurs when a child's blood Pb level reaches 10 microgram per decilitre (μg/dL) in the US, however research has shown Pb to have toxic effects at blood Pb levels less than this regulatory value, causing a loss of several IQ points (Lanphear et al. 2000; Lanphear 2005; Canfield et al. 2003; Jusko et al. 2008). The EPA adjusted the regulation for Pb in floor dust from 100 to 40 μg ft^{-1} (USEPA 2000). Dixon et al. (2009) recently found floor dust Pb values less than 12 μg ft^{-1} to protect the majority of children from having elevated blood Pb (i.e. above 10 μg/dL).

Many factors have been identified as the source of increased levels of Pb in house dust. These were summarized by Fergusson and Kim (1991) and include: distance of home from road, close proximity of home to smelters, mining activity, and/or commercial garages, dustiness and carpet wear, presence of a fireplace, house age, house material, renovations, and redecorations, and occupationally exposed residents. Gaitens et al. (2009) found tobacco to be a significant source of Pb to house dust at low dust loadings. Lead has been identified as accumulating in house dust to concentrations greater than those found outdoors in the immediate environment including garden soil and street dust. A study conducted by Rasmussen et al. (2001) determined a geometric mean of 233 milligrams per kilogram Pb in house dust versus 34 mg kg^{-1} in street dust, and 42 mg kg^{-1} in garden soil in Ottawa, Canada. Similar trends were observed in

Christchurch, New Zealand, Louisiana, United States, and Amman, Jordan (Kim and Fergusson 1993; Lemus et al. 1996; Al-Momani 2007).

Antimony (Sb)

The metalloid Sb has a mean concentration of 0.2 mg kg^{-1} in the earth's crust (Smith and Huyck 1999) and is mainly found in the trivalent and pentavalent oxidation states in environmental media. It is present in the sulphide minerals stibnite and antimonite as well as in the oxide valentinite (Filella et al. 2002). It is associated with silver, Pb, and Cu ores which is of relevance to this study as the city of Greater Sudbury has active Cu mining sites.

Industrially, Sb is used in Pb containing products, such as Pb storage batteries, to increase Pb's physical properties of hardness and strength (Shrivas et al. 2008). In regions where this element is mined, soil samples had greater concentrations of Sb, showing high correlations with As and being readily uptaken by plants (He 2007). This element has obtained much attention in recent years for its use as a catalyst in the manufacturing of polyethylene terephthalate (PET) plastics for water bottles as it can leach into drinking water (Westerhoff et al. 2008). Additionally its use as a flame retardant in a variety of products but especially infant mattresses has caused much debate over its correlation with Sudden Infant Death Syndrome (SIDS) (Richardson 1990; Thompson 1997).

The most toxic form of Sb is the trivalent species which accumulates in the lung, lymph nodes, and hair and has no known biological benefit (Filella et al. 2002). Acute exposure results in nausea, vomiting, and diarrhea (ATSDR 1995), whereas chronic exposure affects blood concentrations of cholesterol and sugar (Westerhoff et al. 2008). On the other hand, Sb has an interesting history in the field of medicine as it has been used to treat a variety of ailments such as fevers, the plague, and syphilis (Smichowski 2008) although it is mainly used in today's society to treat tropical illnesses caused by protozoa (Filella et al. 2002).

Rasmussen et al.'s study (2001) of Ottawa house dust, garden soil, and street dust showed highest concentrations of Sb in house dust versus the latter two media. Interestingly, a Scottish study of contaminated mine soils found Sb to be a low environmental threat since it is relatively immobile and less bioavailable than As or Cu (Flynn et al. 2003).

Yttrium (Y)

Yttrium is a rare earth element with a crustal abundance of 30 ppm (Bottrill 2001). Common uses of this element include camera lenses and televisions (Zhang and Chai 2004) as well as alloys and glass (Bottrill 2001). Health concerns associated with Y include liver damage and lung cancer (Lenntech 2008).

Calabrese and Stanek (1995) recommend Y (as well as aluminum and silicon) to be a reliable soil tracer. This is after many years of studying the ingestion of soil by adults and children (both pica and non-pica children). They have generated formulae to measure soil ingestion by taking into account all items ingested (i.e. food, medicine, soil) and then analysing fecal matter. Yttrium was shown to display low error and have high recovery. This work revealed pertinent information on soil ingestion rates and the percentage of soil in house dust. Based on this work Y is examined in the first paper in this thesis (McDonald et al. 2009a, under review), as a potential tracer of the soil that is tracked into the home on clothes, shoes, and pets specifically in the dwelling entrance.

Zinc (Zn)

The most common Zn minerals include: sphalerite, smithsonite, and zincite (Alcock 1930). Sixty percent of Zn deposits are located in the United States, Mexico, Canada, and

Australia (Mathewson 1959). Zinc is an essential element for good health, however in excess quantities it can lead to nausea and vomiting upon ingestion. Chronic ingestion results in anemia and low cholesterol (ATSDR 2005). Industrially, Zn is used mainly for galvanization purposes but it is also used to produce the alloy brass, dry cell batteries, and is an additive to paint, and in manufacturing rubber and paper (Adriano 2001). Factors influencing Zn concentrations in house dust include presence of a galvanized iron roof on the home, carpet use, make of vacuum cleaner, and presence of rubber carpet underlay (Fergusson and Kim 1991). Several researchers have identified Zn as accumulating in house dust to greater concentrations than found in outdoor soil or street dust (Rasmussen et al. 2001; Turner and Simmonds 2006; Al-Momani 2007). Rasmussen et al. (2008b) found Zn in house dust to be predominately Zn hydroxyl carbonate (~50%), with about 25% each being Zn sulphide and Zn, Fe hydroxides in Ottawa, Canada.

Sources

Outdoor Sources

The three cities involved in this study (i.e. Barrie, Greater Sudbury, and Thunder Bay) all have populations greater than 100 000. Contaminants from mining activities or industrial smokestacks can be spread via wind currents and settle in street dust. Metals can adhere to people's clothes and shoes, and to pets which are then tracked into the home. Outdoor air pollution can also be exchanged with indoor air via indoor/outdoor exchange systems (Komarnicki 2005). A study by Long et al. (2001) suggests that coarser particles like iron have lower infiltration rates into the home from outdoor sources compared to finer particles, like Pb.

A study conducted in a Pb-Zn smelter community in Australia, identified that the presence of such a smelter has caused environmental harm and human health problems. Gulson et al. (2004) found that Pb from this smelter made its way into residential homes via track-in, and then accumulated in dust. They recognized dust as being the most significant contributor to blood Pb. Another study in Nova Scotia, concerning the communities surrounding the Sydney tar ponds, has found that Pb and As from this contaminated site has infiltrated local homes. Analysis of house dust in this area confirmed its Pb and As source to be from the tar ponds and that it was tracked into the home by residents (Lambert and Lane 2004). Harper and Sullivan (1987) identified four metals (Cd, Cu, Pb, Zn) to be higher in concentration in soil closer to industrial waste tips in London and found them to be in even higher concentrations in house dust versus outdoor soil. Similarly Benin et al. (1999) later confirmed that levels of As, Cd, and Pb in house dust in Mexico were highest nearest the industrial activity and decreased with increasing distance from the source. Collectively, these studies confirm that the presence of metals in house dust can originate from industrial outdoor sources and become highly concentrated in residential house dust.

Not all neighbourhoods are surrounded by industrial areas but the majority are accessible by car. Vehicular traffic is another source of metals in house dust. Many studies have identified increased metal concentrations surrounding busy traffic areas. Traffic was named the major source of Cu, Pb, Sb, Zn, barium (Ba), calcium (Ca), cobalt (Co), magnesium (Mg), and titanium (Ti) in house dust in Madrid (de Miguel et al. 1997). Meyer et al. (1999b) studied volume of traffic surrounding homes and concluded that residing near heavily used roads significantly increased the concentrations of As, Cd, and Pb in German house dust. Lower Pb levels in house dust were observed in homes located on cul-de-sacs (Fergusson and Kim 1991; Tong 1998).

The reduction of leaded gasoline usage is a huge success in terms of reducing exposures to this toxic metal which historically was a significant source. Metals are also released from vehicular sources other than gasoline. Fergusson and Kim (1991) noted that distance from roads was a factor influencing the concentrations of Cd, Cu, Pb, and Zn in New Zealand. More specifically, Cu is released from engine wear, thrust bearings, and brake linings, Cr from metal plating, and Zn from motor oils and tire wear (Harrison et al. 1981; de Miguel et al. 1997; Al-Khashman 2004). In summary, the heavy reliance that our society holds on vehicular use can have its consequences.

Climate is another factor with respect to dust accumulation in homes primarily in the form of precipitation and temperature. Precipitation or more specifically humidity has been found to cause increased accumulation of house dust (Park et al. 2000). Thus it is possible for this increased amount of dust to contain more chemical contaminants than dust would in a dry environment. A study by Meyer et al. (1999b) conducted in heavily polluted areas of Germany found that dust levels increased with dampness in the sampled homes. They suggested that increased humidity in the home can encourage particle coagulation and condensation thereby increasing the speed at which contaminants, specifically As, Cd, and Pb, are deposited into house dust. Interestingly, a second study by Meyer et al. (1999a) in the same general area found that dampness was not a significant factor with respect to dust accumulation. Temperature is another important factor in dust contamination levels, as homes tend to become more air tight in times of extreme temperatures. An example of this is keeping windows closed in winter months to conserve energy. This decreases infiltration rates of outdoor sources of metals. Alternatively, it causes increased metal accumulation from indoor sources discussed in the next section due to lack of air exchange (Long et al. 2001). An air tight home also allows for excess humidity to accumulate from events such as cooking and bathing if proper exhaust methods are not functional.

Indoor Sources

Once indoors, heavy metals and other industrial emissions accumulate in house dust due to the abundance of organic carbon in the home. Metals have a high affinity for organic matter and thus metals tend to accumulate in house dust (Rasmussen 2004b). Metals can also contaminate house dust from various activities within the home. Metals are present in many household products and are a component of many hobbies. Thus they are present in the home with the potential of becoming deposited on surfaces.

Ceramics, painting, photography, pottery, and welding are examples of hobbies that involve heavy metals that have the potential to linger and accumulate in house dust. Cadmium from smoking tobacco indoors can play a significant role in adding to the metal content of dust. Tobacco contains approximately 1.8 micrograms per gram Pb, and 1.5 $\mu g\ g^{-1}$ Cd (Willers et al. 1993). Interestingly, however, Willers et al. (1993) found no significant difference between homes of smokers and non smokers with respect to Cd and Pb content of their house dust. Gaitens et al. (2009) however found tobacco use to be a significant contributor to Pb in house dust.

Cooking is also an important source of metals and, unlike specific hobbies listed above, is common to the majority of homes. Frying foods tends to release Al, Ca, Fe, Mg, and Zn (van Winkle and Scheff 2001). One study compared the act of frying bacon and eggs on a gas stove versus being outdoors and found that the cooking event caused a person to be exposed to ten times the amount of small particles that are capable of depositing in the lung (Mitsakou et al. 2007). Candles have also been identified as contributing to the Pb content of house dust. An estimate of 20 percent of the Pb in a candle's wick was identified by van Alphen (1999) to be released into the air and deposited onto floors and furnishings while burning. The Pb in the wick is commonly Pb carbonate, a form of Pb that is easily inhaled and absorbed in the gastrointestinal tract. Another researcher studied candles commercially available in the United States, they were found to release Pb in the range of 0.5 to 66 micrograms per hour and Zn 1.2 to 124 $\mu g\ hr^{-1}$ while burning (Nriagu and Kim 2000).

On a larger scale, home heating can also contribute to metal contamination in house dust. Coal burning has been found to release As (Meyer et al. 1999b). Moriske et al. (1996) included As, Cd, Cr, Pb, and Ni to have higher concentrations in homes heated with coal versus central heating. Lower metal infiltration was identified in homes with HVAC systems, likely due to lower air exchange rates (Long et al. 2001). Since Pb naturally contaminates fossil fuels, gas heated homes may be more susceptible to Pb contaminated dust (Tong 1998).

Products found and used within the home can also contribute to the metal content of house dust. Significant emissions of metals from various household products are listed in van Winkle and Scheff (2001) and include: spot removers and glues (Cu), mothballs (As), and hairsprays (Mg). Carpet backings, rubbers, paints, and fillers are all sources of Zn (Fergusson and Kim 1991). Eye shadows contain metals as colour pigments including As, Co, Cr, Pb, and Ni, these are often not mentioned on the list of ingredients (Sainio et al. 2000). Historically Pb was added to paint and thus older homes potentially are at risk of Pb contamination from chipped paint but more significantly while renovating (Kim and Fergusson 1993). Renovations cause Pb to be released from old paint during its removal (sanding and scraping) and thus Pb becomes available to accumulate in the indoor environment (Lemus et al. 1996).

Meyer et al. (1999a) made the conclusion that in unpolluted areas, indoor activities are the main source of heavy metals in house dust. Caution must be therefore taken to clean up after activities that involve heavy metals or using products containing metals because their release is inevitable. With metals being released from both indoor and outdoor sources, they are abundant in the urban environment. Thorough dust collection (i.e. home cleaning) is thus a necessity to protect the health of all residents within a home.

Thesis Objectives

This thesis will evaluate the wipe method and is the first study to use wipes as the sampling medium in Canada for the purpose of exposure assessment. This study aims to establish baseline values of As, Cd, Cr, Cu, Ni, Pb, Sb, and Zn in urban Canadian homes assessed using wipe sampling. This study is a component of the Canadian House Dust Study (CHDS), Health Canada's national study of which Dr. Pat Rasmussen is Principal Investigator. The current study focuses on the second phase of the CHDS, specifically the cities of Barrie, Greater Sudbury, and Thunder Bay, Ontario. All wipe samples collected in this phase were provided for this thesis by Health Canada.

The limits of detection and quantification for each element will be calculated based on procedural wipe blanks generated in the lab component of this thesis. An important aspect will be to examine the quality assurance samples, both field blanks and collocated duplicate samples. Specifically, variability will be calculated in side-by-side duplicates to assess the reliability and collection efficiency of wipes as the sampling medium.

This study also aims to identify differences in metal loadings in house dust amongst rooms in a home and amongst homes. Industrial emissions and surrounding land use can cause variability in metal loadings amongst homes. Products used within the home can cause variability between rooms in a home. This study will reveal which rooms in a dwelling are most representative of exposures to metals and therefore the best to sample, leading to increased sampling efficiency. Yttrium loadings will be examined due to its potential as a soil tracer. Entry values will be compared to interior rooms to determine spatial associations amongst metals and to estimate the amount of metals that are tracked into the home from outdoor soil.

A final aspect of the study is to propose to Health Canada thresholds for consideration for each metal in indoor floor house dust. Two sets of thresholds will be derived empirically from the data collected in this study, consisting of a lower and upper threshold for each element studied. These two thresholds will define the lower and upper limits of the baseline subpopulation of homes. The upper set of thresholds will identify the elevated group of homes. It is within this group of elevated homes that potential sources of metals can be hypothesized. This information will help risk assessors and risk managers provide Canadians with ways to reduce their exposures to metals in the home.

Study Constraints

Health Canada's Research Ethics Board's ethics protocol to protect the privacy of study participants prevents the data in this thesis from being geocoded. This is why a spatial analysis was not included. Secondly, the goal of the CHDS, of which this study is a part, is to gain a statistically representative sample of house dust from the whole country. Therefore the study was statistically designed for this purpose. Hence, the samples obtained are not representative of individual cities.

Other Contributions

Indoor Guidelines Workshop.

Poster "Wipe Sampling Methodologies to Assess Exposures to Lead and Cadmium in Urban Canadian Homes" included as appendix in: " Guidance Document on Guidelines for Indoor Settled Dust (in prep). By: Wilson Scientific Consulting Inc. and Meridian Environmental Inc. Oct 20th, 2009, Health Canada, Ottawa, ON.

Presentations Given:

Department of Earth Science, University of Ottawa

Presentation on December 4th, 2009. "Metal Toxicity in Urban Canadian Shield Homes Assessed Using Wipe Sampling Methodologies – Review of the Literature and Methodology" Fall 2008 Graduate Seminar Series

Department of Earth Science, University of Ottawa

Presentation September 25th, 2009. "Wipe Sampling Methodologies to Assess Exposures to Metals in Urban Canadian Homes" ISWAN Earth Sciences Seminar Series

Federal Working Group on Children's Environmental Health, Health Canada Presentation

November 12th, 2009. "Wipe Sampling Methodologies to Assess Exposures to Lead and Cadmium in Urban Canadian Homes"

Statement of Originality

The work presented in this thesis was written by myself with crucial input and critical review by Dr. Pat Rasmussen. The wipe digestions were conducted by myself as well as Christine Levesque. The ICP-MS analysis was conducted by Marc Chénier.

Statement of Confidentiality

Due to the confidential nature of this work and the commitment to protect participant privacy, the raw data used in this thesis is not presented in the appendices, in accordance with Health Canada protocols for the Canadian House Dust Study.

Article I.

Wipe Sampling Methodologies to Assess Exposures to Lead and Cadmium in Urban Canadian Homes

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WIPE SAMPLING METHODOLOGIES TO ASSESS EXPOSURES TO LEAD AND CADMIUM IN URBAN CANADIAN HOMES

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ABSTRACT

Wipe sampling methods are widely used to quantify lead (Pb) loadings inside homes. In the present study we expand the wipe sampling method to investigate other elements in addition to Pb, namely cadmium (Cd) and the soil tracer yttrium (Y).

Following the ASTM 1728 sampling protocol, 1372 wipe samples (including field blanks and duplicates) were collected from 222 homes using Ghost Wipes™. All wipe samples were digested according to a modified version of the ASTM 1644 digestion protocol in which hydrofluoric acid was added to enhance extraction efficiency, and analyzed using ICP-MS. Recoveries assessed using NIST certified reference materials were 93±6% for Pb and 88±14% for Cd (n=66).

Results indicated that 43% of Pb and 23% of Cd samples were below LOD (932 ng m^{-2} and 125 ng m^{-2} respectively). Threshold values of 125 μg m^{-2} for Pb and 4.4 μg m^{-2} for Cd, identified using Q-Q plots, were used to distinguish "elevated" loading values from "background" (baseline) loading values. Indoor sources and tracked-in soil were identified as potential contributors to elevated loading values. Spearman ranking indicated strong spatial associations amongst the metals. The study shows that wipe sampling provides useful information on room-to-room variability of metals, shedding light on possible sources of metals in residential environments.

Keywords exposure assessment, housedust, metals, built environment

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1. INTRODUCTION

The average Canadian spends 90% of their time indoors (Health Canada, 2009), and thus there is a growing demand for information on contaminant levels and sources in the indoor environment. In particular, information about sources of metals in the home is necessary to assess childhood exposures caused by ingestion of dust. Industrial land use, traffic emissions, and geological weathering results in the dispersal and settling of metals in soil and street dust which are tracked indoors by residents and their pets. Soil and street dust is thought to contribute anywhere from 20% to 95% of house dust (Rasmussen et al., 2001 and references cited therein). Metals can also originate from interior sources such as paint, household products, crafts and hobbies, and tobacco use, and tend to accumulate in house dust (Rasmussen, 2004).

Cadmium's (Cd) non-corrosive properties allow its application in batteries, pigments, metal coatings, and plastics (Shoeters et al., 2006). At elevated levels of exposure, Cd acts as a nephrotoxicant (Rasmussen and Gardner, 2008 and references cited therein) and as a carcinogen (Bussières et al., 2004). The principal non-occupational exposures to Cd occur through diet (Gamberg and Scheuhammer, 1994; Kormaniki, 2005) and tobacco use (Yapici et al., 2006). A recent risk assessment of Cd in house dust in the Netherlands (Oomen et al., 2008) estimated that the above pathways represent 90% of the total daily intake of Cd, and therefore ingestion of house dust containing Cd could potentially play a significant role in an individual's overall exposure. Cadmium and lead (Pb) differ from essential metals such as copper and zinc which are required by the body in trace quantities for optimal health.

The Canadian government has significantly reduced the Pb content of paint and gasoline over the past few decades (Canada Mortgage and Housing Corporation (CMHC), 2009). Relics of Pb's past abundant use appear during renovations of older homes, potentially increasing Pb exposure (Reissman et al., 2002). The extensive literature on childhood exposures to Pb, pointing to the severe consequences of overexposure on early brain development resulting in learning and behavioral problems, has led to a re-evaluation of the 10 micrograms per deciLitre blood Pb regulatory action value (Lanphear et al., 2000; Canfield et al., 2003; Lanphear et al., 2005). Hornung et al. (2009) identified the need to maintain low Pb exposure throughout childhood as blood Pb at 6 years of age was found to be more highly correlated with neurological health effects than at 2 years of age.

Presently, there are no Canadian guidelines for metal loadings in indoor residential dust. The United States Environmental Protection Agency (USEPA) has set a regulation for Pb in house dust at 40 $\mu g\ ft^{-2}$ on floors, based on the collection of house dust using the wipe method (USEPA, 2000). For the purpose of determining individual exposures to residential sources of Pb, many researchers view wipe sampling as superior to vacuum sampling. Wipe sampling mimics a child's hand contact with hard surfaces and it is thus argued that a child has greater potential to be exposed to metals in dust settled on smooth surfaces versus carpeted surfaces (Yiin et al., 2002). Additionally, Rodes et al. (2001) found that only one third of a child's hand actually touches a surface, therefore vacuum sampling may not represent tangible exposure. The question of where to sample was addressed by Wilson et al. (2006) as USEPA (2000a) does not specify the optimal location or number of samples to be taken within the home. Wilson et al. (2006) identified the best predictor of elevated blood Pb in a child to be non-carpeted floor wipe samples collected in the home's entry area, living room, kitchen, and bedrooms.

The purpose of the present study is to quantify metal loadings in urban homes with the aim of establishing background levels for residential exposure in Canada. As such it represents the first published dataset of its kind for Canada. The only other published Canadian study to use wipes as the sampling medium was conducted by CMHC (1995) to test cleaning methods for Pb in paint dust. The present study also aims to identify differences in Pb and Cd loadings between rooms, thereby providing insight on possible sources. The entry of the home is a prime location to study metals that infiltrate the home from outdoor sources. Similarly, interior rooms such as kitchens, living rooms, and bedrooms are more likely to be influenced by indoor sources of metals. Ultimately, the information presented here will assist in quantifying typical Canadian exposures to Pb and Cd, and reveal ways to reduce exposures.

2. MATERIALS AND METHODS

2.1 Selection of Wipe Brand

Out of the many brands of wipes available on the market, the Ghost Wipes™ brand was selected for use in this study. Based on preliminary testing at Health Canada (Rasmussen, 2007a unpublished data) Ghost Wipes™ were determined to be optimal because they meet all criteria in the American Society for Testing and Materials (ASTM) method E 1792 (2002a); they completely digest in acid (consistent with previous findings by Harper et al., 2002); and they contain very low background concentrations of the elements in this study. Ghost wipes™ also meet criteria outlined by Millson et al. (1994) in their evaluation of market available wipes including robustness, high precision, and ease of use.

2.2 Sampling and Analysis

A total of 1372 wipe samples were collected from 222 homes between October 2007 and March 2008 in three Ontario cities: Barrie (57 homes), Greater Sudbury (86 homes), and Thunder Bay (79 homes). Homes were randomly selected as part of a sampling strategy designed for a larger nation-wide study (Rasmussen et al., 2007b), and therefore the sampling was not intended to be representative of individual cities. The present study incorporates all samples collected in the 2007-2008 sampling season. The collection of information by sampling technicians using questionnaires and interviews, and the communication of results and guidance to the participants, is described elsewhere (Rasmussen, 2007b).

Up to ten wipe samples were collected from each home, from smooth surfaces in the middle of each room. Rooms that were sampled included: main entry, kitchen, living room, family room, adult's bedroom, child's bedroom, and child's primary play area, based on guidance provided by Wilson et al. (2006). Other rooms were occasionally included on an *ad hoc* basis. A total of 932 different rooms were sampled using wipes, yielding 932 individual wipe measurements plus 440 quality assurance measurements (total = 1372 wipes).

Wipe samples were collected according to ASTM E 1728 protocol (2002b), which prescribes a vertical and horizontal overlapping S-shaped movement applying even pressure to the floor surface. The collected wipe was folded inward to preserve the sample and placed directly into a labeled plastic digiPREP™ digestion tube which was sealed inside Ziploc™ bags for transport to the lab. The wipes were shipped to Health Canada, Environmental Health Centre, Ottawa Ontario, Canada, and stored frozen until time of analysis.

The analytical method employed in this study was based on a modification of ASTM method E 1644 (2004), a nitric acid digestion. The modifications consisted of (1) adding hydrofluoric acid during digestion to increase digestion efficiency, (2) multi-element determination by ICP-MS, and (3) incorporating the use of a digiPREP™ heating block. The goal of adding hydrofluoric acid was to quantify total metals in the dust (i.e. maximize recovery). It is noted that the human gastrointestinal tract is estimated to be capable of absorbing less than 30% of the total Pb in house dust (Turner and Ip, 2007).

2.3 Quality Assurance and Quality Control

A 12 square inch plastic template was used to constrain the wipe sampling area, as prescribed by the ASTM E 1728 protocol (2002b). The template was cleaned with an alcohol wipe between rooms and a new template was used for each home. In each home sampled, one field blank wipe and one field duplicate wipe were collected. The field blank wipe was exposed to all handling procedures used for the samples with the exception that no surface was wiped (ASTM, 2002b). The room from which the duplicate was collected rotated amongst homes. The sampling strategy yielded a quantity of blanks (n=220) and duplicates (n=220) in excess of the minimum frequency of 5% as outlined in the ASTM E 1728 protocol (2002b), incorporating a high proportion of quality assurance data into the study design.

Throughout sample digestion, three procedural reagent blanks and three procedural wipe blanks were included per batch (n=37 batches). Three certified reference materials for Pb and Cd were included: NIST 2583 indoor dust, NIST 2584 indoor dust, and NIST 2711 Montana soil (certificates do not include yttrium). The mean recovery and standard deviation of these certified reference materials was 93 ± 6% (n=66) for Pb and 88 ±14% for Cd (n=66).

The limits of detection (LOD) and quantification (LOQ) for each element were calculated based on three times and ten times the standard deviation of the lab procedural wipe blanks respectively (n=110). For sample results less than the LOD, half the LOD was substituted where required. The detection limits for Pb, Cd, and yttrium (Y) were calculated to be 932 ng m^{-2}, 125 ng m^{-2}, and 9.09 ng m^{-2} respectively.

2.4 Data Analysis and Units

SPSS® Statistics v. 17.0 and Microsoft Excel® (2007) with the Analyse-it add-in (version 2.20) were used for statistical analyses. Shapiro-Wilks tests of the datasets resulting from this study revealed non-normal distributions, and therefore non-parametric statistical methods were employed. Spatial relations were investigated using Spearman rank correlation coefficients, and 50^{th} and 95^{th} percentiles were used to summarize Cd and Pb loadings within rooms and within homes.

To convert from SI units (μg m^{-2}) used in the present study, to units of μg ft^{-2} (microgram per sq ft) used in the USA, multiply loading values expressed in μg m^{-2} by a factor of 0.0929.

3. RESULTS AND DISCUSSION

3.1 Wipe Data Quality

Field blank and collocated duplicates were collected from all but two homes in this study (n=220). The LODs, LOQs, and medians and 95^{th} percentiles of the field blanks are reported for each element in Table 1.

Table 1. Limits of detection (LOD) and limits of quantification (LOQ) for Cd, Pb, and Y are based on 37 analytical batches (three procedural wipe blanks per batch). Results for field wipe values (median; 95th percentile) include all field wipe blanks collected in this study (n=220).

	LOD (ng m^{-2})	LOQ (ng m^{-2})	Field Wipe Blank (ng m^{-2}) Median; 95th Percentile
Cd	125	416	< LOD; 444
Pb	932	3110	< LOD; < LOD
Y	9.09	30.3	< LOD; < LOD

The relative percent difference (RPD) between collocated duplicate samples was calculated using the equation RPD = $((dup_2 - dup_1)/((dup_2 + dup_1)/2)*100)$. Note that the mean RPD in the range between LOD and LOQ is greater than the mean RPD above LOQ (Table 2). The greater variability in the lower range (between LOD and LOQ) reflects a combination of field and analytical sources of uncertainty, and points to the need to consider the more rigorous LOQ as the appropriate criterion for quality assurance.

Table 2. Mean Relative Percent Difference (RPD) of collocated duplicates categorized by limits of detection (LOD) and quantification (LOQ). Total number of pairs = 220. See Table 1 for LOD and LOQ values.

	Duplicates <LOD	Between LOD and LOQ		Duplicates >LOQ	
	No. of pairs	No. of pairs	Mean RPD	No. of pairs	Mean RPD
Cd	53	103	51.3	64	39.4
Pb	100	40	71.8	80	41.2

Results for Pb showed that 382 samples were below LOD, which equals 43% of the total of 932 wipe measurements. For Cd, 196 samples were below LOD, or 23% of the wipe measurements. In the case of Y, 34 samples were below LOD (or 3.6% of the measurements). With respect to the LOQ, 511 samples (55%) were below the LOQ for Pb, 672 (72%) were below LOQ for Cd, and 106 (11%) were below LOQ for Y (n=932).

There were a total of 36 homes in which all Pb loading values, in all rooms, were below LOD. In the case of Cd, there were 11 homes in which all samples were below LOD. This yielded a subset of 186 homes for Pb, and 211 homes for Cd, in which at least one wipe sample exceeded LOD. It is this subset of homes which is subjected to further analysis and interpretation in the discussion below.

3.2 Lead Loadings

Figure 1 is a normality (Q-Q) plot of the maximum Pb loading observed in each of the subset of 186 homes having at least one wipe greater than the LOD. Note that the majority of the data fall on the line representing a lognormal distribution, with exceptions occurring at the extreme high and low ends. That portion of the dataset which falls on the lognormal line is considered the "background" subpopulation, for the purpose of this paper. Eight points occur above the breakpoint in the high end of the dataset, where the values start to trend away from the lognormal line (Figure 1). This breakpoint occurs at about 125 $\mu g\ m^{-2}$ (or 12 $\mu g\ ft^{-2}$). Three homes within the elevated subpopulation had wipe samples exceeding the USEPA (2000a) regulation for Pb in floor dust, i.e. 40 $\mu g\ ft^{-2}$ or 431 $\mu g\ m^{-2}$.

The upper breakpoint in the Q-Q plot at 125 $\mu g\ m^{-2}$ (or 12 $\mu g\ ft^{-2}$). is used herein as an empirical threshold to distinguish between "background" and "elevated" Pb loading subpopulations. This selection of a threshold at about 12 µg ft^{-2} is consistent with a recent US residential study which concluded that Pb loadings less than 12 µg ft^{-2} should be protective for the majority of children (Dixon et al., 2009).

Out of the eight above-threshold homes in Figure 1, three homes displayed the highest Pb loading in the entry way. In two homes the highest Pb loading occurred in an adult bedroom; in two homes the highest Pb loading occurred in children's bedrooms; and in one home, the highest loading occurred in a child's play room.

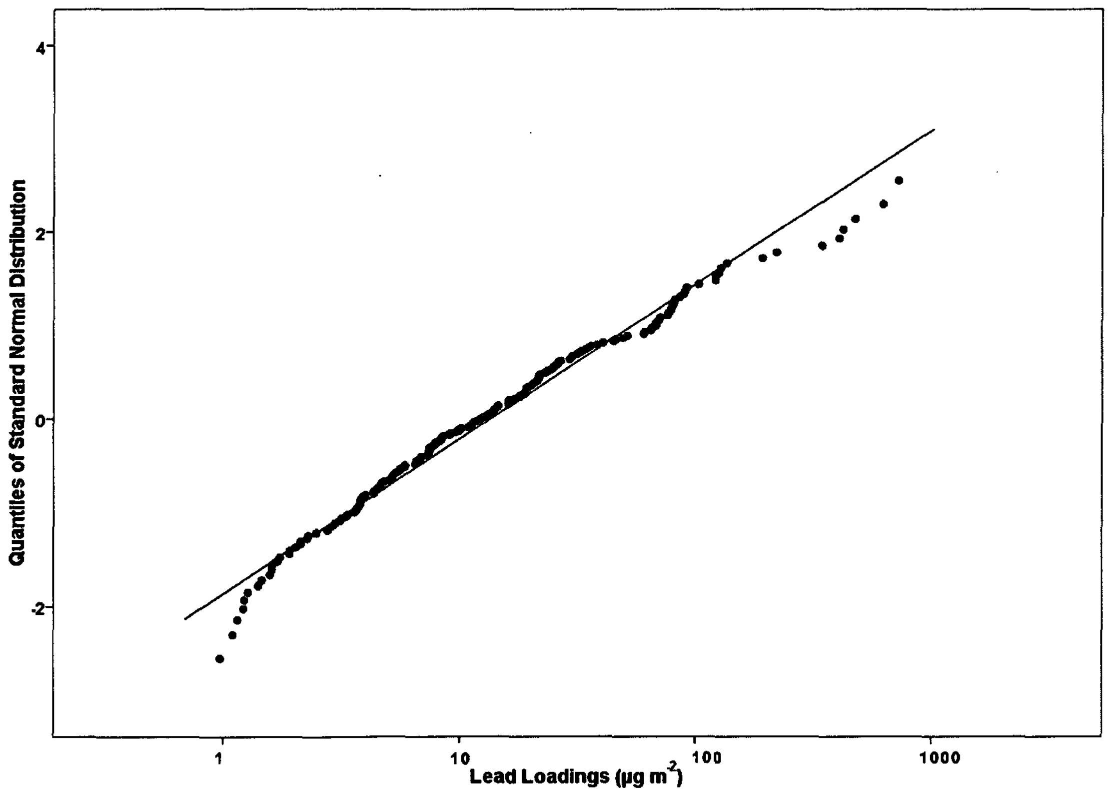

Figure1. Normality Plot (Q-Q plot) for log transformed Pb loadings occurring in a subset of homes having at least one wipe above the limit of detection (n=186). Homes are represented by the wipe with the highest Pb loading, regardless of the room from which it was taken. The line

through the data represents a lognormal distribution. Eight homes are above the breakpoint in the dataset which occurs at 125 μg m^{-2}.

3.2.1 Sources of Lead

A room by room analysis was conducted with the aim of exploring potential sources of Pb (Table 3). Overall, the entry way displayed the highest median value however other noteworthy rooms include adult bedrooms and children play rooms. Notably, the highest individual value was located in a child's bedroom. Lead loadings in the home's entry suggest outdoor sources such as track–in of dirt, whereas elevated loadings in bedrooms and play rooms suggest interior sources of Pb. It is concluded from the results shown in Table 3 that both indoor and outdoor sources of Pb contribute to Pb loadings in house dust.

Table 3. Summary of Pb loadings (μg m^{-2}) by room for all 222 homes sampled.

(LOD = 0.932 μg m^{-2}).

Room	n	Percent Below LOD	50th Percentile	95th Percentile	Maximum value
Entry	208	22	5.64	87.5	619
Kitchen	218	49	1.02	21.2	165
Living / Family room	114	49	< LOD	40.9	485
Adult Bedroom	93	43	3.26	75.9	422
Child Bedroom	50	60	< LOD	133	720
Play room	23	26	3.50	66.9	220

Age of the home appears to be an important factor, as six out of the eight homes above threshold were built before 1960. The mean age of the eight above-threshold homes is 1954 ± 29 years, which is (on average) nineteen years older than homes in which all wipe samples were below LOD (1973 ± 22 years; n=36). A review of the literature indicates that Pb-based paint is most commonly cited as the primary cause of elevated Pb in older homes (Rasmussen, 2004).

Potential sources of metals in the above-threshold homes may be hypothesized based on questionnaire responses. Residents of two of the above-threshold homes were employed in jobs where Pb is used or encountered, for instance on mining and shipyard sites. In one of these homes, Pb is stored in the house for craft and hobby use. A resident of another home habitually conducts bodywork on vehicles in the driveway: the entry wipe for this home displayed both elevated Pb and Cd loadings. Recent renovations involving painting (four homes) and plumbing (one home) were reported to have occurred in some homes with elevated Pb loadings. This may be relevant as renovation activity in older homes can increase Pb availability where high Pb content paint has been used in the past (CMHC, 2009). Sampling technicians noted that old paint was chipping off the wall in one home with elevated Pb loading built before 1960. Two homes with high Pb loadings reported that occupant(s) smoke indoors, which may be relevant as Pb from tobacco use has been indicated as a source of Pb on interior surfaces (Gaitens et al., 2009).

3.3 Cadmium Loadings

A normality (Q-Q) plot for Cd loadings (Figure 2) indicated a breakpoint in the dataset at about 4.4 $\mu g\ m^{-2}$ (0.4 $\mu g\ ft^{-2}$). There were nine homes in this study with Cd loadings above this threshold value: three of these were kitchen wipes, three were entry wipes, two were office wipes, and one was from an adult bedroom.

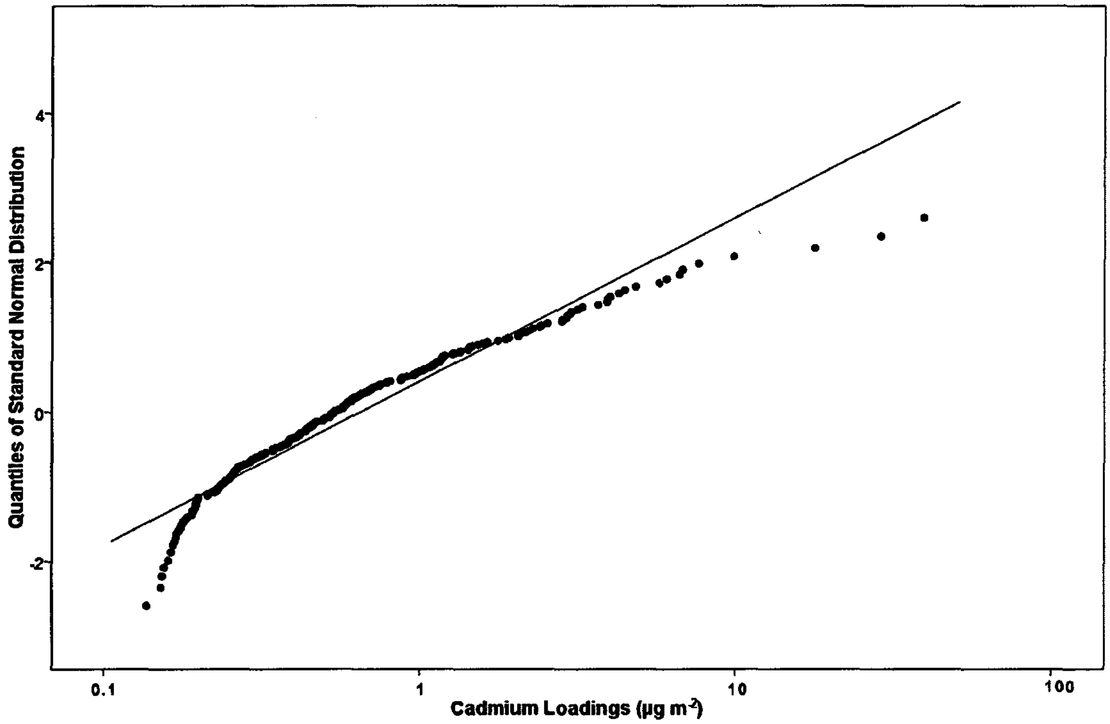

Figure 2. Normality Plot (Q-Q plot) for log transformed Cd loadings occurring in homes having at least one wipe above the limit of detection (n=211). Homes are represented by the wipe with the highest Cd loading, regardless of which room it was collected from. The line through the data represents a lognormal distribution. Nine homes occur above the breakpoint in the dataset which occurs at 4.4 $\mu g\ m^{-2}$.

3.3.1 Sources of Cadmium

As in the case of Pb, there are both indoor and outdoor sources of Cd. Cadmium loadings are relatively high in home entry ways and adult bedrooms compared to other areas of the home (Table 4). As exterior sources of Cd are associated with both industrial land use and geological sources, track-in of dirt by residents and their pets is a plausible explanation for the observation of relatively high loadings in entry ways. Higher Cd loadings in adult bedrooms versus child occupied rooms suggest tobacco use as a possible source. Questionnaire data indicated that 14%

of homes in the study were occupied by at least one person who smoked inside the home. House age may also be a factor: the mean age and standard deviation of homes with Cd loadings above threshold was 1962 ± 29 years (n=9), which is a thirteen year age difference compared to homes where all wipe samples were below LOD (1975 ± 23 years; n=11).

Table 4. Summary of Cd loadings (μg m^{-2}) by room for all 222 homes sampled (LOD= 0.125 μg m^{-2}).

Room	n	Percent Below LOD	50th Percentile	95th Percentile	Maximum value
Entry	208	16	0.314	3.11	8.14
Kitchen	218	28	0.207	1.24	9.84
Living / Family room	114	20	0.239	1.20	5.66
Adult Bedroom	93	22	0.276	2.74	19.0
Child Bedroom	50	28	0.199	1.68	3.27
Play room	23	17	0.320	1.06	1.63

Information gathered from the questionnaire results revealed clues about possible sources in the nine homes with above-threshold Cd loadings. In three homes where painting was listed as a hobby, elevated Cd loadings were observed in the room where this hobby was conducted. Crafts and hobbies are known to have the potential to influence metal loadings in the home (Rasmussen, 2004), as Cd is a common component of certain paint pigments (Harte et al., 1991). In four other homes activities were reported that involve metal work such as welding, soldering, and body-work on vehicles. These activities are potential sources, as Cd is a common component of solders and fluxes (Harte et al., 1991). Of the two remaining elevated homes, one contained a resident who smokes and one had recent plumbing and painting renovations. Five of these nine homes above threshold were built before 1960.

3.4 Metal Correlations

Spearman correlation coefficients (r_S) were calculated to compare Cd, Pb, and Y loadings for various rooms within the home. With regard to the entry wipe samples (n=208; Table 5) correlations between Pb and other metals were greater than r_S = 0.5, which are strong relationships according to definitions by Reimann et al. (2008). No significant differences were observed for analyses of individual locales compared to analyses of the entire dataset. Correlations were similarly strong for bedroom and other interior wipe samples (i.e. kitchens, living rooms, bedrooms, and playrooms; Table 5). Since all correlations were strong, information about precise sources of these metals could not be determined from this type of analysis.

Calabrese and Stanek (1995) determined Y to be a reliable soil tracer for use in the estimation of soil ingestion rates. Yttrium is a rare earth element with an average concentration of 30 ppm in the earth's crust (Bottrill, 2001). The comparison of Y against Cd, and Pb in wipe samples was included in Table 5 in an effort to identify the relative contribution of outdoor and indoor sources of these metals in different areas of the home (Table 5). The highest proportion of soil is likely to be found in dust samples collected in the home's entry areas, due to track – in of outdoor dirt by residents and their pets.

The results in Table 5 are inconclusive as to whether indoor or outdoor sources dominate. Strong Pb – Y correlations (r_S = 0.7) and moderate to strong Cd – Y correlations (r_S = 0.6) are found for wipes collected in entry ways (Table 5). However, correlations are equally strong for wipes collected in bedrooms and other interior rooms (Pb-Y r_S = 0.6, Cd-Y r_S = 0.6). These results suggest that both indoor and outdoor sources exist for all three elements.

Yttrium has limited use in household products: it is generally alloyed in small amounts with other metals, and is most commonly found as the oxide yttria (Y_2O_3), used for making red phosphors in colour television picture tubes (Chemistry Encyclopedia 2007; Bottrill 2001). In summary it appears that Pb, Cd, and Y are contributed to house dust from both indoor and outdoor sources, based on the correlations in Table 5 observed in all entry and interior subsets.

Table 5. Spearman rank correlation coefficients for Cd, Pb, and Y sub-divided by room. "Interior room wipes" include living rooms, kitchens, bedrooms, and play rooms.

Location	n	Cd-Y	Pb-Cd	Pb-Y
Entry wipes	208	0.6	0.7	0.7
Bedroom wipes	189	0.6	0.7	0.6
Interior room wipes	575	0.6	0.6	0.6

4. CONCLUSIONS

This research has generated the first multi-element wipe sampling database for "background" or baseline urban residential environments in Canada. The information obtained by applying the wipe methodology to Canadian residential environments assists in quantifying typical urban residential exposures to Pb and Cd, and reveals valuable information about variations in metal loadings amongst individual rooms within homes.

The results indicate that activities conducted in each room and the products used within them contribute to the metal level of that room. Similarly, outdoor sources contribute to differences in metal loadings of homes due to track-in of outdoor dirt by residents and their pets.

The finding of strong correlations for Y against both Pb and Cd in entry areas as well as interior areas confirms that both indoor and outdoor sources are important for these three elements.

Ninety-nine percent of homes in this study fell below the regulation of 40 μg ft^{-2} (431 μg m^{-2}) for Pb in floor dust (USEPA, 2000a). The finding that only a small percentage of homes had a wipe sample that exceeded the USEPA regulation (3 out of 222 in total) is consistent with US residential studies such as NHANES (National Health and Nutrition Examination Survey) which reported that the geometric mean of Pb in floor dust was 1.1 μg ft^{-2} (12 μg m^{-2}; Dixon et al., 2009). In the present study, analysis of the maximum wipe loading per home using Q-Q plots suggested thresholds of 125 μg m^{-2} for Pb and 4.4 μg m^{-2} for Cd, which are used to distinguish between background and elevated subpopulations. This dataset will contribute to the development of guidance for reducing exposures to residential Pb and Cd that is specific to the Canadian urban environment.

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Article II.

Extending Wipe Sampling Methodologies to Elements Other Than Lead

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Extending Wipe Sampling Methodologies to Elements Other Than Lead

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Abstract

Wipe sampling is the USA regulatory protocol for measuring "dust lead loadings" in residential environments. Few studies have applied the wipe sampling method to metals other than lead (Pb) for the purpose of residential exposure assessments. This study was undertaken to develop and expand the wipe method for quantifying additional metal(loid)s including arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), and antimony (Sb); and to provide information on typical baseline loadings for these metals in urban Canadian homes.

A total of 932 wipe samples, 220 field blanks, and 220 duplicate wipes were collected from 222 homes located in three cities in Ontario, Canada. Wipe samples were digested according to a modified version of the USA protocol for Pb, which prescribes a nitric acid/hot plate digestion and analyzed using Inductively Coupled Plasma – Mass Spectrometry (ICP-MS). The key modification was the addition of hydrofluoric acid to improve recoveries of the target metals.

Results were analysed with the goal of distinguishing homes characterized by elevated metal loadings from homes characterized as "background", by deriving an upper background threshold for each element using normality (Q-Q) plots. Potential sources of metals in the subset of elevated homes (above-threshold) were interpreted on the basis of their spatial distribution within the homes, combined with information derived from questionnaires. This study demonstrates that wipes are an appropriate sampling option to determine multi-element loadings in residential environments.

Keywords wipe sampling · lead · arsenic · cadmium · chromium · copper · nickel · antimony · dust · residential risk assessment

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Introduction

Wipe sampling is the regulatory method for sampling lead (Pb) in house dust in the United States for the purpose of exposure assessments (USEPA 2000a; HUD 2001). Many researchers have identified the wipe method as suitable for quantifying Pb loadings in residential house dust (Sterling et al. 1999; Farfel et al. 1994; McDonald et al. 2009, under review), and it is the established method for health physics applications (Rasmussen 2004). Few studies however, have used the wipe technique to measure residential loadings of metals other than Pb. Estimates of indoor exposures to metal(loid)s such as arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), antimony (Sb), and zinc (Zn), which are routinely required for residential risk assessments, currently rely on dust concentrations, such as the vacuum dust, soil, and street dust dataset provided by Rasmussen et al. (2001).

The term loading refers to an area measurement, reported in units of mass per surface area (e.g. $\mu g\ m^{-2}$). This differs from a concentration measurement which is reported as mass of contaminant per mass of dust (e.g. $\mu g\ g^{-1}$). Residential loading studies are scarce to non-existent for metals other than Pb, and hence the first objective of the present study is to evaluate data quality and report limits of detection (LOD) and quantification (LOQ). The distribution of the resulting datasets are then examined with the purpose of identifying threshold loading values to assist in distinguishing elevated loadings from background loadings, for each of the target metals.

The ultimate goal of the present study is to provide baseline information on metal loadings in typical urban Canadian homes, for elements other than Pb including As, Cd, Cr, Cu, Ni, Sb, and Zn. Previous studies by Lioy et al. (1992) and Freeman et al. (1997) used wipes to measure Cr in "hotspot" regions where elevated exposures were associated with local industrial sources. In contrast, the present study does not focus on hotspots, but aims to quantify baseline loadings in randomly sampled urban residential neighbourhoods. This study also investigates spatial trends in metal loadings within homes, comparing the main entry areas with interior rooms to assist in interpreting potential outdoor and indoor sources.

Materials and Method

Sampling Procedures

The samples used in this study were collected from 222 randomly selected urban homes as part of a larger multi-phase national baseline study (Rasmussen et al. 2007). Specifically this subset includes all samples collected from January 2008 to March 2008 (second phase of the national study) from three cities in Ontario Canada: Barrie, Greater Sudbury, and Thunder Bay. Participants were asked not to clean their home one week prior to the technician's visit. Upon arrival, technicians completed a questionnaire with the help of the resident, according to study protocols described elsewhere (Rasmussen et al. 2007). Information collected in the questionnaire included: age of home, recent renovations, and occupations and hobbies of residents.

The Ghost Wipes™ brand was chosen for use in this study. The wipe is a nonwoven polyvinyl alcohol fibre pre-moistened with distilled water (Boeniger 2006). This product meets all criteria listed in the American Society for Testing and Materials (ASTM) method E 1792 (2002a) with the additional advantage that the Ghost Wipes™ brand dissolves completely in acid (Harper et al. 2002). Carpeted floors were not sampled, which resulted in variations in the number of samples collected per home. In the homes sampled an average of 56% of floors were covered in carpeting while 44% were non-carpeted and thus potentially available to be wipe sampled.

Rooms that were sampled included: main entry area, kitchen, living and family rooms, bedrooms (both adult and child), and children's play areas. From these rooms, 706 wipe samples were collected. Additional rooms were sampled on an *ad hoc* basis as per the discretion of the sampling technician. The samples from these additional miscellaneous rooms totalled 226, yielding 932 wipe samples altogether. An average of 4 wipe samples were collected per home, with a maximum of 10 and a minimum of 1, excluding the 2 quality assurance (QA) wipes collected per home.

A 30 cm square plastic template was used to constrain the wipe sampling area, as prescribed by the ASTM E 1728 protocol (2002b). Accordingly, wipe samples were collected using (1) a horizontal S-shaped pattern through the template, followed by (2) a vertical S-shaped pattern, with (3) focus on each of the four corners. As far as possible, the middle of each room was selected to collect fresh dust, as recommended by Wilson et al. (2006). The collected wipe was then folded inward to preserve the sample and placed directly into a labelled plastic digiPREP™ digestion tube which was then sealed inside a Ziploc™ bag for shipment to the lab.

The wipes were received at Health Canada, Environmental Health Centre, Ottawa Ontario, Canada, where they were stored frozen until time of analysis.

Analytical Procedures

In preparation for analysis, wipes were digested using a modification of ASTM method E 1644 (2004). The first modification consisted of adding 5 mL hydrofluoric acid (to 10 mL concentrated nitric acid) during digestion to increase digestion efficiency. This is to address the problems reported by Hassan et al. (2007) related to low recoveries for elements that occur in refractory compounds. The second modification involved substitution of a digiPREP™ heating block for a traditional hot plate. The digiPREP™ features programmed automatic shut-off based on monitoring both temperature and volume of solution. The final modification was the dilution of samples to 50 mL rather than 100 mL to avoid diluting the sample digest below instrument detection limits for elemental analysis.

Background contamination within the Ghost Wipe™ (n=51) was quantified during method development. Mean and standard deviation (μg per wipe) of the elements in this study were 0.3 ± 0.02 for As; 0.1 ± 0.1 for Cd; 1.3 ± 0.4 for Cr; 1.0 ± 0.1 for Cu; 0.03 ± 0.6 for Ni; 1.2 ± 0.6 for Pb; 0.4 ± 0.002 for Sb; and 15.5 ± 2.4 for Zn. Due to the high Zn background, Ghost Wipes™ were determined to be not suitable for the quantitation of Zn in house dust.

Every effort was made to avoid sample contamination throughout the study. In the field, sampling was performed wearing powder-free nitrile gloves, and wipes were placed directly into digiTUBES™ where they remained sealed until digestion in the digiPREP™ heating block. Digestion proceeded within a metal-free High Efficiency Particulate Air (HEPA) filtered polypropylene constructed fume hood. Ultrapure reagents were used including: hydrofluoric acid (47-51% w/w; Seastar Chemical Inc., Baseline, Sidney, B.C., Canada), nitric acid (69-70% w/w; Baker Instra Analysed, Reagent Grade, Phillipsburg, N.J., U.S.), hydrogen peroxide (30% w/w; EMD Chemicals Inc., Suprapur, Darmstadt, Germany), and distilled, deionized water (18.2 MΩ cm). A cover remained over the auto sampler throughout analysis to prevent contamination of the samples and standards. These combined efforts minimized contamination, which was monitored using reagent blanks and procedural lab wipe blanks, and improved detection limits.

The digests were analysed at Health Canada using a Perkin Elmer Sciex Elan DRC II Inductively Coupled Plasma Mass Spectrometer (ICP-MS), equipped with a cross-flow nebulizer, Ryton Spray Chamber, platinum skimmer and sampler cones. The instrument control software is Elan Version 3.4. An eight point calibration curve was calculated from the concentrations listed: 0, 10, 30, 50, 100, 500, 1000, 2500 parts per billion (ppb) and was matrix matched following USEPA 200.8 protocols (USEPA 1994). An internal standard consisting of 25 ppb each of germanium, indium, and rhenium was used to correct for interferences.

Quality Assurance and Quality Control

QA wipes were collected in 220 of the 222 study homes, consisting of one field blank wipe and one field duplicate wipe, both collected in the same room, totalling 440 QA wipes. This sampling strategy yielded > 20% field blanks and > 20% duplicates, which is in excess of the 5% recommended in the ASTM E 1728 protocol (2002b), to incorporate a high proportion of QA data into the study design. The duplicate wipe was taken adjacent to the room sample, and the room location from which QA samples were collected (i.e. kitchen, living room etc.) was rotated amongst homes. The field blank wipe was exposed to all handling procedures used for the samples with the exception that no surface was wiped (ASTM 2002b).

A total of 37 analytical batches were required to digest the samples and QA wipes. An average of three procedural reagent blanks, three procedural wipe blanks, and two certified reference materials CRM-spiked wipes were included per batch. National Institute of Standards and Technology (NIST) CRMs were rotated amongst: NIST 2583 indoor dust (n = 24), NIST 2584 indoor dust (n = 23), and NIST 2711 Montana soil (n = 23). The two NIST indoor dust CRMs were collected by vacuuming homes in the U.S. and sieving with a 100 μm and 90 μm sieve (NIST 2583 and 2584 respectively). NIST 2584 was collected preferentially from homes involved in lead poisoning intervention programs. To prepare CRM-spiked wipes, a Ghost Wipe™ was unfolded on a clean surface and a specified mass of NIST CRM was placed on the centre of the wipe. The wipe was then folded inward and placed in a digiTUBE™ for digestion. A mass of 0.1 g was used for NIST 2583 and NIST 2711, and a mass of 0.015g for NIST 2584 to account for differences in the Pb concentrations of the CRMs. ICP-MS results for the digestion of the CRM-spiked wipes were used to calculate recoveries for each of the metals.

The LOD and LOQ for each element were calculated based on three times and ten times the standard deviation of the lab procedural wipe blanks respectively (n=110) in accordance with USEPA protocols (2000b). For sample results less than the LOD, half the LOD was substituted where appropriate.

Data Analysis

SPSS® Statistics v. 17.0 and Microsoft Excel® (2007) with the Analyse-it® add-in v. 2.20 were used for statistical analyses. Each dataset was determined to have non-normal and non-lognormal (with the exception of Cr) frequency distribution by Shapiro-Wilk tests, and therefore non-parametric statistical methods were used. Spearman rank correlation coefficients were calculated for the elements to assist in interpreting spatial trends. In the entry and interior rooms' discussion, the 226 samples from extra rooms were not included in the room – to – room variability analysis (entry and interior wipes). In cases where two entry ways (or any other duplicate room) were sampled, the room with the lower Pb loading was excluded so that a given room type was only counted once per home. The standard deviation (SD) of duplicate samples is reported to assess variability in side-by-side samples. The equation to calculate SD from duplicates (from Gluer et al. 1995) is:

$$SD = \sqrt{\sum_{j=1}^{m} d_j^2 / 2m}$$

where m is the number of duplicate pairs, and d is the difference between duplicates. To convert from SI units (μg m^{-2}) used in the present study, to units of μg ft^{-2} (microgram per square foot) used in the USA, multiply loading values expressed in μg m^{-2} by a factor of 0.0929.

Results and Discussion

Recoveries

Recoveries were calculated from CRM-spiked wipes (Table 1). Generally Cr, Cu, Ni, and Sb had better results in dust CRMs (NIST 2583 and 2584) than in the soil CRM (NIST 2711). Recoveries for Cu (59-72%) were lower than observed for the other elements; As and Ni were slightly better (74-92% and 67-75% respectively); and Cr, Cd, Pb, and Sb displayed highest recoveries (78.8-95.7). In terms of precision, the relative standard deviation (RSD) was less than 20% in all cases except Cd in NIST 2583, Cr in NIST 2711, and Ni in NIST 2711. Lead displayed the smallest relative standard deviation for all three CRMs and was less than 10% in each case.

Freeman et al. (1997) used a patented sampling device: "LWW dust sampler" which uses pre-weighed filters rather than Ghost Wipes™. They additionally used sulphuric acid and ICP-AES and were able to report a higher recovery of 98.6% for Cr from NIST samples. The Freeman et al. (1997) study was optimized for a single element, Cr, while the current study was optimized for a suite of elements, including As, Cd, Cr, Cu, Ni, Pb, and Sb, which requires the lower detection limits of ICP-MS.

Table 1 Recoveries (%) of elements using wipes spiked with certified reference materials; NIST 2583 (n=24), NIST 2584 (n=23), NIST 2711 (n=23).

Element	**NIST 2583**	**NIST 2584**	**NIST 2711**
As	91.6	73.9	75.4
Cd	93.7	86.4	90.7
Cr	82.8	81.5	78.8
Cu	n/a	72.2	58.7
Ni	n/a	75.0	66.7
Pb	90.2	92.6	95.7
Sb	n/a	90.8	88.8

Determination of Acceptance Criteria

Limits of detection and quantification (LOD and LOQ) are reported for each element in Table 2 along with the percentage of wipe samples which yielded results below these limits. The medians of the field blanks are also included and each was less than the respective LOD for each element. As indicated earlier, Zn is not included in this analysis due to unacceptably high background concentrations indicated by the wipe blanks (15.5 ± 2.4 μg per wipe).

Table 2 Limits of detection (LOD) and quantification (LOQ) for the elements in this study, calculated for 37 analytical batches (approximately three procedural wipe blanks per batch). The number of wipes less than LOD and LOQ are calculated as a percentage of the total number of wipe samples collected (n=932). Median and 95th percentile values for field blanks (n=220) have been wipe blank-corrected.

Element	LOD (μg m^{-2})	% < LOD	LOQ (μg m^{-2})	% < LOQ	Field Blank Wipe Median; 95th %ile (μg m^{-2})
As	0.15	55	0.51	66	< LOD; < LOD
Cd*	0.13	23	0.42	72	< LOD ; 0.44
Cr	0.90	10	3.01	29	< LOD; 0.45
Cu	4.06	77	13.5	87	< LOD; < LOD
Ni	1.19	41	3.97	63	< LOD; < LOD
Pb*	0.93	43	3.11	55	< LOD; < LOD
Sb	0.18	41	0.60	68	< LOD; < LOD

from McDonald et al. (2009, under review)

With respect to the collocated duplicate wipes, standard deviation (SD) is presented in the context of LOD and LOQ values for each element in Table 3. Note that for each element duplicate variability is lower for duplicate pairs within the category between LOD and LOQ compared to the category above LOQ (Table 3). The higher duplicate variability of the QA wipes above LOQ (Table 3) most probably reflects the heterogeneity of house dust, and the small area sampled (a 30 cm square area). Higher loading values can result from spot sources such as a paint chip that may occur in the small area sampled, with the result that higher loadings will display greater variability using side-by-side duplicates. The r^2 value is higher for each element in the category above LOQ than between LOD and LOQ (see Table 3 for the number of pairs in each category). Above LOQ, the regression is significant for all elements except Cu which is the element with the lowest r^2 value and also the lowest number of paired duplicates above LOQ (Table 3). In the category between LOD and LOQ the r^2 values are lower for each element and none are significant (all p-values greater than 0.05). The standard deviations of the collocated duplicates revealed greater variability above LOQ than between LOD and LOQ.

Table 3 Standard Deviation (SD) of collocated duplicates categorized by limits of detection (LOD) and quantification (LOQ). Total number of pairs = 220. See Table 2 for LOD and LOQ values.

Element	No. of Duplicate Pairs < LOD	Variability between LOD and LOQ			Variability Above LOQ		
		No. of pairs	SD	r^2 (p-value)	No of pairs	SD	r^2 (p-value)
As	120	28	0.41	<0.01 (0.74)	72	2.47	0.55 (<0.001)
Cd	53	103	0.09	0.03 (0.069)	64	5.28	0.57 (<0.001)
Cr	24	64	1.63	0.01 (0.51)	132	13.6	0.64 (<0.001)
Cu	172	27	13.8	<0.01 (0.90)	21	128	0.05 (0.36)
Ni	98	72	2.07	<0.01 (0.72)	50	50.2	0.79 (<0.001)
Pb	100	40	1.32	<0.01 (0.75)	80	19.3	0.71 (<0.001)
Sb	91	61	0.21	0.04 (0.11)	68	1.17	0.43 (<0.001)

Distinguishing the Baseline Subpopulation of Homes

To evaluate the overall dataset, and determine threshold loading values, each home was represented by its maximum wipe value. The goal of this analysis is to distinguish homes characterized by elevated metal loadings from homes characterized as "background", by defining the upper limit of "background" for each element using normality (Q-Q) plots. This analysis was done separately for each element, using the maximum wipe result for each home regardless of the identity of the room yielding the maximum result. Homes in which all wipe samples were less than LOD for any given element were disregarded in this analysis, as the goal is to define an upper threshold, or the upper limit of the background subpopulation. The number of disregarded homes ranged from 1 to 110 homes, depending on the element (individual sample sizes are reported in Table 4).

An upper threshold for Cd (4.4 μg m^{-2}) and Pb (125 μg m^{-2}) were previously determined from normality (Q-Q) plots of the log-transformed maximum wipe value per home (McDonald et al. 2009, under review). In the case of Pb, the empirically derived threshold coincides with that of a recent national study conducted in the U.S. where 12 μg ft^{-2} (approximately 125 μg m^{-2}) was determined to be the threshold below which children should be at minimal risk of elevated blood Pb levels (Dixon et al. 2009; McDonald et al. 2009, under review).

Similarly, Q-Q plots of log-transformed data were prepared for the elements addressed in the present study. Nickel is shown in Figure 1 and it is representative of As, Cd, Cu, Pb, and Sb where an upper breakpoint is distinguishable in each of the datasets. The exception is Cr where there is no observable breakpoint: instead, the data does not trend away from the line, indicating a lognormal distribution (Figure 1). Note that the majority of the data for As, Cu, Ni, and Sb also fall on the line representing a lognormal distribution, with exceptions occurring at the extreme high and low ends. As described in McDonald et al. (2009, under review) the upper breakpoint serves as a useful threshold to define the group of homes that would be considered "elevated" in loadings for a given element. That portion of the dataset which falls on the lognormal line is considered the "background" or baseline subpopulation, for the purpose of this paper.

Establishing the breakpoint is a subjective decision, especially in the case of As where there is less space between data points at the breakpoint. In this case, 6 homes were included in the elevated group of homes based on the selection of the break at the point where the homes begin to trend away from the lognormal line. The breakpoint is more easily selected in the case of Cu, Ni, and Sb. No breakpoint was observed for Cr as it is lognormally distributed as indicated both by the Q-Q plot and by the Shapiro-Wilk test ($p = 0.754$). Therefore, in the case of Cr the homes with the six highest Cr loadings (Figure 1) are selected for review of potential sources.

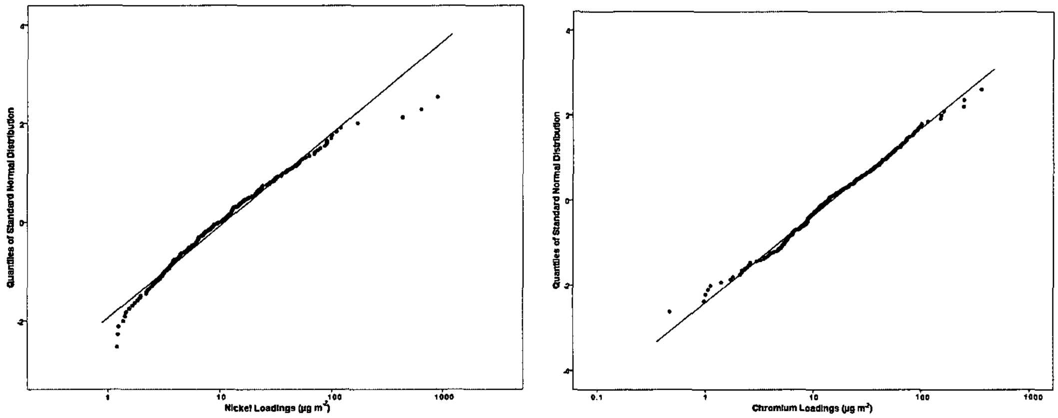

Fig. 1 Normality (Q-Q) plots for Ni loadings (which is representative of the other 3 elements: As, Cu, Sb) and Cr loadings (the only element that is lognormally distributed) in homes having at least one wipe above the limit of detection. Homes are represented by the wipe with the highest loading, regardless of the room from which it was taken. The line represents a lognormal distribution in each case. See Table 4 for the numerical value of the breakpoint and the number of homes above threshold for each element.

Using the above-described technique, a subset of 28 homes having at least one element (As, Cd, Cu, Ni, Pb, and Sb) above threshold was isolated for further analysis. The number of homes that occur above the breakpoint for each element is listed in Table 4 along with the numerical value of the upper threshold.

On average, the subset of homes above the upper thresholds (irrespective of the element) is fourteen years older than the group of homes where all wipes collected were less than LOD. Note that house age does not appear to be a factor for some of the elements (e.g. Ni). House age was examined due to the fact that paint historically had high Pb content (CMHC 2009) and because it is widely cited in the literature as a factor resulting in higher dust Pb concentrations

(Rasmussen et al. 2001; Sutton et al. 1995). Results of this analysis suggest that older homes may not only be at risk of elevated Pb loadings but other metals as well. This is supported in the literature where paint on an 80 year old New Orleans home contained varying quantities of many of the metals in this study: 9.7g Cd, 2.4g Cr, 14.8g Cu, 1.5g Ni, and 7.4kg Pb (Mielke et al. 2001).

Table 4 Age (mean ± sd.) of homes separated into two categories.

Element	n	Age (mean ± sd.) of Homes With 100% of Wipes < LOD	n	Age (mean ± sd.) of Homes > the Upper Thresholds	Upper Loading Thresholds (μg m^{-2})
As	61	1971 ± 26	6	1964 ± 32	25
Cd*	11	1975 ± 23	9	1962 ± 29	4.4
Cr	1	1993 ± 0	0	n/a	n/a
Cu	110	1969 ± 30	5	1967 ± 20	200
Ni	46	1980 ± 25	4	1982 ± 18	125
Pb*	36	1973 ± 22	8	1954 ± 29	125
Sb	36	1971 ± 24	3	1984 ± 9.0	10

*from McDonald et al. (2009, under review)

Potential Outdoor Sources

As mentioned earlier, the Cr dataset is lognormally distributed and therefore an elevated subset of homes could not be identified using the breakpoint method. However, examination of the six homes with the highest Cr loadings (Figure 1) reveals interesting information. All six of these homes had the highest Cr loading in the entry area. This observation points toward outdoor sources of Cr as dominant contributors to house dust. Heavy vehicular traffic was reported in front of 100% of these homes, 67% were located either near mining or pulp and paper industrial activity, and 50% were near railways. All of these homes reported high daily traffic of residents and all but one home had either a cat or dog that went in and out of the home on a daily basis.

Additionally, occupations where Cr may be encountered were reported in four homes and include: construction, mechanic, shipyard worker, and two residents from one home work on a mine site. These observations suggest that Cr may be adhering to resident's shoes and clothes from outdoor sources and subsequently tracked into the home where it is deposited primarily in the entry.

While estimates of the contribution of soil to house dust vary anywhere from 20 to 95% in the literature (Rasmussen et al. 2001 and references cited therein), the USEPA supports an upper estimate of 70% (USEPA 1998). This large range is probably a reflection of the heterogeneity of house dust. It may be assumed that the primary entry to the home reflects outdoor sources of metals due to track–in from the outdoor environment on clothes and shoes. Risk of soil ingestion by a child's normal hand–to–mouth activity is therefore likely greatest in the home's entry way. Results from the questionnaire data indicate that in 61% of the elevated subset of homes, there is high traffic in and out of the home on a daily basis by residents and their guests. Metals can also be brought home by employees who work with or encounter metals on their job site. Occupations of residents from homes above the set of upper thresholds that may fit this description include: artist, construction worker, firefighter, miner, roofer, and welder.

Exterior land use as a factor for higher metal loadings in the home was addressed by the questionnaire. For the subset of homes above the upper thresholds, (n=28) heavy traffic was noted for 79% of these homes. Various researchers around the world have cited heavy vehicular traffic to be a cause of elevated As, Cd, Cu, Pb, and Sb in house dust (Fergusson and Kim 1991; de Miguel et al. 1997; Meyer et al. 1999). Industrial activity is another potential source of elevated residential metal loadings which has been well investigated (Benin et al. 1999; Gulson et al. 2004; Lambert and Lane 2004). In the present study, proximity to mining activity was noted in the questionnaire for 39% of homes above the upper thresholds. Railways were noted to be of close proximity (within 2 km), in 25% of homes above the upper thresholds, irrespective of the element.

Entry Wipes

An entry wipe was collected in 208 of the 222 homes (14 homes with carpeted entrances were not sampled). Elements that display higher median values from the entry wipe, compared to interior room wipes, include: As, Cd, Cr, Ni, Pb, and Sb (Table 5). While the median value for both interior and entry wipe samples for Cu are below LOD, the same trend of increasing loading from interior rooms to entry is observed in the 95th percentile and maximum values, where there is an increase from 20.1 to 109 and 198 to 1200 μg m^{-2}, respectively. Copper loadings are characterized by the widest range, having the greatest percentage of wipes below LOD yet also having 95th percentile and maximum values exceeding the other elements studied. Among entry wipes Cr had a remarkably low quantity of wipes below LOD followed by Cd.

Spearman rank correlation coefficients (r_s) were calculated for the entry subset, (n=208) (Table 6, top row). A correlation greater than or equal to 0.5 is strong according to definitions by Reimann et al. (2008). The analysis revealed strong correlations in the entry for every metal combination with the exception of Ni-Sb. A similar correlation matrix was calculated by Al-Khashman (2007) for street dust samples however, Pearson's correlation coefficient was used. The author reported similarly high correlations for Cu-Cr = 0.85, Cu-Pb = 0.72, and Cr-Ni = 0.69.

Spearman correlations amongst the elements in interior rooms (n=498) were generally less strong than in the entry (Table 6, bottom row). Overall, the correlations for As, Cr, Cu, Ni, and Sb are stronger in the entry than interior rooms. This complements the findings in Table 5 where higher 50th and 95th percentiles as well as maximum values were observed for the entry versus interior rooms. Collectively, the two statistical approaches suggest that these elements (As, Cr, Cu, Ni, and Sb) have greater contributions from the outdoor versus indoor environment, reflecting Al-Khashman's findings. The interior room correlation between Cd-Pb (0.61) indicates a contribution of Cd and Pb from indoor sources.

Table 5 Summary of entry loadings (µg m^{-2}) for each element (n=208) listed first, followed by summary of interior loadings (µg m^{-2}) for each element (n=498), underneath. "Interior rooms" include living rooms, family rooms, kitchens, bedrooms (adult and child), and play rooms.

Element	Wipe Location	Percent < LOD	50th Percentile	95th Percentile	Maximum Value
As	Entry	40	0.49	15.1	39.0
	Interior	60	< LOD	7.25	19.8
Cd*	Entry	16	0.31	3.11	8.14
	Interior	24	0.23	2.23	19.0
Cr	Entry	3.4	10.4	86.9	364
	Interior	12	5.08	33.3	77.8
Cu	Entry	61	< LOD	109	1200
	Interior	84	< LOD	20.1	198
Ni	Entry	27	4.43	70.2	900
	Interior	42	1.65	16.6	98.6
Pb*	Entry	22	5.64	87.5	619
	Interior	48	1.25	61.0	720
Sb	Entry	26	0.49	4.77	23.1
	Interior	48	0.21	2.74	17.8

*from McDonald et al. (2009, under review)

Table 6 Spearman rank correlation coefficients (r_s) of entry wipes (n=208) listed first, followed by interior room wipes (n=498). "Interior rooms" include living rooms, family rooms, kitchens, bedrooms (both adult and child), and play rooms.

Element	Wipe Location	As	Cd	Cr	Cu	Ni	Pb
Cd	Entry	0.50					
	Interior	0.35					
Cr	Entry	0.61	0.58				
	Interior	0.54	0.49				
Cu	Entry	0.52	0.62	0.64			
	Interior	0.23	0.44	0.39			
Ni	Entry	0.61	0.49	0.60	0.70		
	Interior	0.29	0.44	0.48	0.46		
Pb	Entry	0.50	0.70*	0.65	0.73	0.64	
	Interior	0.42	0.61*	0.51	0.47	0.55	
Sb	Entry	0.52	0.64	0.55	0.52	0.44	0.64
	Interior	0.36	0.47	0.54	0.38	0.49	0.52

*from McDonald et al. (2009, under review)

Potential Indoor Sources

Questionnaire information was used to identify potential sources of metals. For example, homes with loadings of analysed elements above the upper thresholds had a mean age older than homes where all wipe samples taken were less than LOD for As, Cd, Cu, and Pb (Table 4). Within the group of homes above the upper thresholds (n=28), 43% had been recently painted. One of these 12 homes was elevated in every element. Of the remaining 11 homes, five had high Pb loadings, three homes had high Cd loadings, two homes had high As, and one home had high Ni. Recent plumbing renovations were reported in three homes, of these, one home was elevated in both Cd and Pb, one had high Cu, and the third had elevated Sb. An above threshold Sb loading was recorded in one home, where carpet removal occurred during wipe sampling. Antimony is commonly used as a flame retardant in household furnishings and carpets (Filella et al. 2002). Renovation activities, such as painting and plumbing, can result in elevated exposure to metals, most notably Pb (Roberts et al. 2009) due to the past high Pb content of paint (CMHC 2009). A higher level of cleaning may be required in older homes to minimize dust levels thereby reducing exposures to metals (Roberts et al. 2009).

The activities and hobbies of residents as well as the products they use, contribute to metal loadings of interior rooms and thus should be examined for potential interior sources. Eight homes in the above upper thresholds category (n=28) had occupants who reported painting as a hobby. Of these homes, three were above the Cd threshold, two were above the Ni threshold, and one home each was above the As, Pb, and Sb thresholds. One home was above threshold in all elements (As, Cd, Cu, Ni, Pb, and Sb), as displayed by the entry wipe. Possible sources may be associated with car repairs, including bodywork and welding, conducted in the driveway of this home. Soldering was an activity reported in three homes, two of which had elevated Cd loading while the third had an elevated Ni loading. One occupant in the elevated subset of homes reported making stained glass windows at home (which typically involves Pb); however, it was not Pb but As in this home that was above the upper threshold. An occupant of another above-threshold home makes Pb sinkers for fishing use. The activities in these two cases were not reported to occur inside the home however it was the entry wipe in these two homes which had the highest loading.

Tobacco smoking was reported in 14% of homes in both above-threshold homes (n=28) and in the total study (n=222), suggesting there is no significant difference in metal loadings between smoking and non-smoking homes. In contrast Gaitens et al. (2009) concluded that tobacco use is a source of Pb in house dust on interior surfaces. Crafts and hobbies such as those discussed in the preceding paragraphs have been identified as potential sources of metals within the home (Rasmussen 2004; Harte et al. 1991). Similar home Pb-work has been correlated with elevated childhood exposure via house dust in Japan (Kawai et al. 1983). Despite the usefulness of the questionnaire to suggest potential sources, it should be noted that Gallicchio et al. (2002) found questionnaires to be less reliable than the numerical wipe loadings with regard to information on potential Pb hazards in the home.

Interior Wipes

A total of 498 interior wipes were collected in the study, from kitchens, living rooms, family rooms, bedrooms and play rooms. Summary statistics for these rooms are reported in Table 5 (bottom row). In each case the 50th and 95th percentiles are lower than for entry wipes (Table 5). This sheds light on the relative contribution of metals from the two environments (both indoor and outdoor). The interior room maximum values for Cd and Pb are the only two statistics in Table 5 where the interior loading is greater than the entry loading. This supports the assumption that metals (As, Cd, Cr, Cu, Ni, Pb, Sb) enter the home from outdoor soil and contribute to house dust in the primary dwelling entry. The lower values of As, Cr, Cu, Ni, and Sb in interior rooms suggest fewer indoor sources of these metals. However the higher maximum value for Cd and Pb in the interior room versus entry suggests that there are substantial sources of Cd and Pb in the indoor environment, causing the elevated values. Therefore it is likely that indoor sources of Cd and Pb are important contributors to loadings in some of the homes in this study. Reduction of use of these sources could play an important role in managing exposures to these metals.

Conclusions

Two types of thresholds were identified for each element in this study. The first type was a quality assurance threshold obtained from the calculation of a limit of detection (LOD) and limit of quantification (LOQ) for each element. Values above the LOQ showed greater measurement uncertainty than values between LOD and LOQ, based on the standard deviation of collocated duplicate wipes.

The second type of threshold identified homes that could be categorized as having elevated metal loadings compared to the baseline subpopulation. An upper threshold could be identified for each element with the exception of Cr. These upper thresholds were interpreted from the Q-Q plots of the log-transformed value for the maximum wipe per home. Questionnaire data from the subset of homes above the upper thresholds (n=28) provided limited information about potential sources of elevated loadings of elements in the wipe samples.

The elements studied (As, Cd, Cr, Cu, Ni, Pb, Sb) accumulate in house dust from both indoor and outdoor sources. The data obtained in this study suggest that higher percentages of soil occur in entry way dust while lower percentages occur in dust in interior rooms. This study revealed room-to-room variability with generally greater loadings observed in the entry for all elements. The exceptions were Cd and Pb which displayed higher maximum values for interior rooms as compared to the home's entry. By sampling a variety of rooms in each home, pertinent information on sources specific to different rooms within the home was available. This is an advantage of wipe sampling, which permits investigations of room-to-room variability. A disadvantage of the wipe method is the large number of sample results below LOD for the analyzed elements. Additionally, loadings above LOQ displayed high variability as determined using collocated duplicate wipes.

The present study revealed potential sources of the study elements and hence has provided information on ways to reduce exposures in the urban environment. Ultimately, it is important to maintain low dust levels within the home to reduce exposures to metals. Wipes have been shown to be a useful method for collecting multi-element loading data (i.e. As, Cd, Cr, Cu, Ni, Pb, Sb). Ghost Wipes™ were found to be not suitable to measure Zn loadings, due to the high background Zn concentrations. It is anticipated that wipe sampling for a wide range of elements, in addition to Pb, will prove to be a useful measure of exposure for future residential risk assessment and risk management decision-making.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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Discussion and Conclusions

The work presented in this thesis has identified that Canadians are exposed to metals via their house dust and that the degree of these exposures varies between rooms within a home and between homes. The dust in a dwelling's entry way reflects outdoor sources with soil being a dominant contributor. Dust located in interior rooms reflects the products used within these rooms with less contribution from soil. Since this is the first study to use wipes for exposure assessment in Canada the objective was to collect a large quantity of wipes per home. Up to ten wipes were collected in each home. However, the exact number varied due to the presence of carpeting. Rooms that were sampled as per the CHDS sampling instructions included the entry, kitchen, living room, family room, adult's bedroom(s), child's bedroom(s), and child's play room. Samples from other rooms totalled 226, examples include: craft room, dining room, home office, hall, music room, sitting room, and sun room.

To minimize cost and increase efficiency for future studies the number of samples collected per home could be decreased. Based on observations and general trends of the data collected in this study, samples from 3 to 4 key rooms would provide an adequate representation of the whole home. These critical rooms include the entry, living room and / or child's play room, and child's bedroom. These rooms represent daily exposures however if a resident conducts a hobby (where metals are a component) in a specific room an elevated loading may be observed there.

To minimize exposures to metals the universal advice is simply to keep dust levels in the home to a minimum. This study has identified many potential sources of metals including various crafts and hobbies as well as products that may contribute to a resident's exposures.

Wipe sampling was advantageous to use in this study due to its ability to show room-to room variability.

Limitations of Wipe Sampling

Not all researchers prefer to use the wipe method. Several disadvantages of using wipes as the sampling medium were observed in conducting this research. A large percentage of wipes were less than the limit of detection for each element. Chromium had the smallest percentage less than the LOD followed by Cd, whereas Cu had the highest followed by As. Additionally, there was high variability when collocated duplicate wipes had numeric loadings below the limit of quantification.

Lichtenwalner (1992) refers to wipe sampling as an imprecise technique. This statement is based on the fact that in his study he wiped the same surface twice and found detectable metal concentrations on the second wipe. On average, 55% of the metals detected on the first wipe were present on the second wipe leading to his disproval of the sampling methodology. Wipes are often constrained to sampling hard non-porous surfaces. When wipes were tested on porous surfaces such as plywood, their removal efficiency was as low as 43% (Chavalitnitikul and Levin 1984). Wipes often tear on rough surfaces and their robustness is thus important in choosing a good wipe. Another constraint is the presence of aloe on wipes as these wipes were identified as having high background Pb content and their use should thus be avoided (Millson et al. 1994).

Another point of weakness of the wipe sampling technique is the question of how much pressure to apply during sampling. It is important to keep the pressure applied by the technician's hand constant during all sampling to obtain comparable data. One strategy is to designate a sole person to the sampling which was the method used in the current study. However a more precise technique was developed and patented, the "Lioy-Weisel-Wainman"

(LWW) sampler (Lioy et al. 1993). This method involves a filter being placed on a rectangular device which is then used to wipe the designated area. Placing a hand on the device versus the wipe evens out the pressure applied by the sampler (Lioy et al. 1993; Adgate et al. 1995). The LWW sampler has been tested by several researchers and is referred to as a successful sampling device. Freeman et al. (1996) found it to be an efficient field instrument when measuring Cr on windowsills. The LWW sampling device was also found to have a significantly higher reproducibility (between paired samples) for Pb loadings ($r = 0.87$) versus the HUD wipe method ($r = 0.71$) (Rich et al. 1999). The LWW sampler has added benefits in that it collects less substance from cracks in surfaces and it provides a concentration value (mass contaminant per mass dust) in addition to the loading value (mass contaminant per surface area) (Lioy et al. 1993).

The choice of sampling medium ultimately depends on the purpose of the study. Wipes are useful for exposure assessments where the flooring is non-carpeted while vacuuming is a good way to sample carpets. Yet vacuuming has been the traditional technique in dust collection thus many researchers have attempted to compare the two techniques with side by side sampling to test the validity and efficiency of wipe sampling.

Comparison of Wipe versus Vacuum Sampling

Some experts in the field argue that carpets are a better surface to sample due to their tendency to trap and collect dust versus hard surfaces where contaminants are free to move around and accumulate unrepresentatively (Kim and Fergusson 1993). Lanphear et al. (1995) compared two vacuum methods with wipe sampling and found highest Pb loadings using wipes. The authors indicated that wipes were superior to vacuuming due to ease of use and cost, concluding that wipes are better for large scale sampling. Sterling et al. (1999) conducted an experiment with three vacuum methods as well as the HUD wipe method and found one vacuum method, the high-volume small surface sampler (HVS3) to exceed the other two vacuum methods as well as the wipe method. This was evaluated in terms of greatest to lowest dust collection. In spite of this, their conclusion identified wipe sampling to be superior even for carpeted surfaces. In contrast Reynolds et al. (1997) compared wipe methods with vacuuming and found that vacuuming was more efficient on carpeted surfaces. They were less assertive in their conclusions, neglecting to state which sampling method best estimated contamination in house dust. However both sets of researchers did claim that wipes were easier to use and more cost effective. Similarly, research by Farfel et al. (1994) supports wipe sampling as they found that the HUD vacuum method is likely to underestimate Pb loadings in comparison with the wipe method. Wipes were used in the current study solely on non-carpeted surfaces, while vacuuming was conducted on carpeted surfaces.

Both sampling methods have their advantages and limitations. The most important consideration in choosing a sampling method is the purpose of the study. Wipes are an excellent method for exposure assessment.

Impacts of Thesis to Risk Assessment and Policy

This research was partly funded by Health Canada's Vulnerable Populations Division, to fill a data gap noticed by the Federal Working Group on Children's Environmental Health. This group became aware that wipes are the regulatory method in the United States to test children's exposures to Pb in house dust. As such there is extensive data available with respect to Pb loadings in homes in the United States. However, they were unable to compare this data with Pb in Canadian homes as dust has been sampled in the past by vacuuming (mass contaminant per mass dust) in Canada. Wipes (mass of contaminant per unit surface area) have not been used in Canada for the purpose of exposure assessment. The Vulnerable Populations Division therefore provided funding to obtain such a dataset to identify the quantity of Pb in urban Canadian homes, and to compare this data with loadings observed in the United States.

The work presented in this thesis was also valuable to Health Canada's Dust Guidelines Workshop, which took place on October 20^{th}, 2009, in Ottawa, ON. A paper copy of the poster of this research entitled "Wipe Sampling Methodologies to Assess Exposures to Lead and Cadmium in Urban Canadian Homes" (Conference on Contaminated Soil, Sediment, and Water, Oct 19-22, 2009) was provided at this workshop (Appendix A). The group is reviewing dust sampling methods (vacuum versus wipe) to be implemented in Canada. The poster will be included in their forthcoming report entitled "Guidance Document on Guidelines for Indoor Settled Dust" as an appendix.

The Canadian Mortgage and Housing Corporation (CMHC) is also interested in this research project. As Canada's national housing agency they are concerned with Pb in the Canadian housing stock. Their website contains many documents on Pb hazards in the home with ways to reduce exposures to this toxic element. Their website recommends the use of wipes to test for Pb in house dust, providing the procedure to correctly take the sample and advice to the public on where to send the sample(s) for analysis. Previously this organization released a study using wipes in Canada where the purpose was to test cleaning methods for Pb in paint dust (CMHC 1995).

Lastly, this research is part of the Canadian House Dust Study which received mention as Exhibit 2.5 in the "Report of the Commissioner of the Environment and Sustainable

Development to the House of Commons" released by the Office of the Auditor General of Canada (2009). The exhibit on the study emphasized the importance of quantifying contaminants in the built environment. This study will provide risk assessors and risk managers with information on the amount and type of contaminants to which typical Canadians are exposed.

Influence of Geology

The cities of Greater Sudbury and Thunder Bay are both located in the Canadian Shield; Barrie however is located in Paleozoic geology. Differences in trends were observed between the two settings as demonstrated in the graphs in Appendices B and C of the 95^{th} percentiles for the different rooms sampled. In general greater loadings were observed for Cd, Cu, Ni, and Pb in the Shield versus Barrie. The same trend was observed for As and Cr except that kitchen loadings were slightly higher in Barrie. Interestingly, Sb loadings are approximately equivalent in Barrie and the Shield homes. House age is a confounding factor. The mean age of homes located in the Shield is twenty six years older than the mean age of homes in Barrie. It should be mentioned once again that the study design was not intended to statistically represent individual cities. Consequently, it should not be interpreted that homes on the Shield are older than homes in Barrie from this thesis.

Recommendations for Future Research

Since this research was conducted as part of the larger CHDS where the purpose was to obtain a statistical representation of the entire country, it would be interesting to add extra sampling locations in the three cities in this thesis so as to be able to statistically represent each city. A spatial analysis of variability between homes would then be possible to confirm the role of geology and land use.

A second issue would be to find an appropriate tracer. The issue with a loading value, obtained by wipes, is that it does not differentiate between the amount of metal per mass of dust, like vacuuming does. That is to say that there may be a wipe with a lot of dust but a small quantity of metals or a wipe could have little dust but a high quantity of metals. Previous to this study Rasmussen (2004b and references cited therein) found that aluminum and barium were elevated in garden soil as compared to indoor dust. However, Rasmussen (2004a) lists several products in which these elements are found and therefore act as indoor sources. Calabrese and Stanek (1995) found yttrium to be a reliable soil tracer. It was therefore examined in McDonald et al. (2009a, under review) however high correlations between Y-Cd and Y-Pb in entry and interior rooms suggest that there are both outdoor and indoor sources of yttrium and it is therefore not a good tracer to use. Future work could explore using uranium as a tracer.

Conclusions

This research has generated the first multi-element wipe sampling database in Canada. This study has successfully shown that the Ghost Wipe^{TM} is appropriate for sampling not only Pb but also As, Cd, Cr, Cu, Ni, Sb, and Y. It has also shown that there is not a single room that is best to sample within a home. This is in part due to the heterogeneous nature of dust as there are many sources (both indoor and outdoor) that contribute to dust levels. Room – to – room variability was demonstrated successfully by the wipe method which allowed for the identification of potential sources of metals.

Thresholds distinguishing the background subpopulation from elevated homes were empirically derived from normality (Q-Q) plots of the dataset for each element studied. The exception to this was Cr as it followed a log-normal distribution and thus no breakpoint in the dataset occurred. The 125 $\mu g\ m^{-2}$ threshold for Pb, observed in this study, converts to 12 $\mu g\ ft^{-2}$

which is the same as the loading rate recommended to protect children from elevated blood Pb levels (Dixon et al. 2009). The consistency of these two studies supports the method used for selecting an upper threshold.

This manner of analysing the data can be extended to other elements which have not been studied as extensively as Pb. The other thresholds are as follows: As = 25 μg m^{-2}, Cd = 4.4 μg m^{-2}, Cu = 200 μg m^{-2}, Ni = 125 μg m^{-2}, Sb = 10 μg m^{-2}. These thresholds may be useful to risk assessors to develop guidelines for these metals in indoor floor dust. Risk assessors and risk managers may also use the information gathered in this study about potential sources to help the public reduce their exposures to these elements. Due to the amount of time Canadians spend indoors, it is essential to be aware of the sources of metals within the home to maintain minimal exposures.

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Appendix A

WIPE SAMPLING METHODOLOGIES TO ASSESS EXPOSURES TO LEAD AND CADMIUM IN URBAN CANADIAN HOMES

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ABSTRACT

Wipe sampling methods are widely used to quantify lead (Pb) loadings inside homes. In the present study we expand the wipe sampling method to investigate other elements in addition to Pb, namely cadmium (Cd) and the soil tracer yttrium (Y).

Following the ASTM 1728 sampling protocol, 1372 wipe samples were collected from 222 homes using Ghost Wipes™. All wipe samples were digested according to a modified version of the ASTM 1644 digestion protocol in which hydrofluoric acid was added to enhance extraction efficiency, and analyzed using ICP-MS. Recoveries assessed using NIST certified reference materials were 93±6% for Pb and 88±14% for Cd (n=66).

Results indicated that 43% of Pb and 23% of Cd samples were below LOD (932 ng m-2 and 125 ng m-2 respectively). Threshold values of 125 μg m-2 for Pb and 4.4 μg m-2 for Cd were identified based on breakpoints in the data on the respective normality (Q-Q) plots. Indoor sources and tracked-in soil were identified as potential contributors to elevated values. Spearman ranking indicated strong spatial associations amongst the metals. The study shows that wipe sampling provides useful information on room-to-room variability of metals, shedding light on possible sources of metals in residential environments.

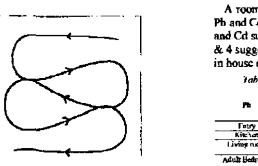
Keywords: exposure assessment, housedust, metals, built environment

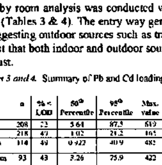
1. OBJECTIVES

Presently, there are no Canadian guidelines for metal loadings in indoor residential dust. The USEPA has set a regulation for lead (Pb) in house dust at 40 μg ft-2 on floors, based on the collection of house dust using the wipe method (USEPA, 2000). The purpose of the present study is to quantify metal loadings in urban homes with the aim of establishing background levels for residential exposure in Canada. As such, it represents the first published dataset of its kind for Canada. The present study also aims to identify differences in Pb and cadmium (Cd) loadings between rooms, thereby providing insight on possible sources. Ultimately, the information presented here will assist in quantifying typical Canadian exposures to Pb and Cd, and reveal ways to reduce exposures.

2. MATERIALS AND METHOD

- This study comprises 222 homes from 3 Ontario cities which were randomly selected as part of a sampling strategy designed for a larger nation-wide study (Rasmussen et al., 2007a).
- Samples were collected from each home's entryway, kitchen, living room, family room, adult's bedroom, child's bedroom, and child's play room provided they were not carpeted (Wilson et al. 2006, Rasmussen et al., 2007a).
- Ghost Wipes™ were chosen as the sampling medium as they meet ASTM 1792 standards (2002a) and dissolve completely in acid (Rasmussen et al. 2007b; Harper et al., 2002).
- Sampling procedure followed ASTM 1728 (2002b) "S shaped" movement within one square foot template in the centre of rooms.
- One duplicate and one field blank were taken from each home which exceeds the QA/QC outlined in ASTM 1728 (2002b).
- Samples were digested in acid following a modified version of ASTM 1644 (2004) where HF was added to HNO_3 to increase extraction efficiency. Sample digests were then analyzed by ICP – MS.
- The limit of detection (LOD) and quantification (LOQ) for each element were calculated based on 3 and 10 times the sd of the lab wipe blanks respectively (n=110). For all values less than LOD, half the LOD was substituted.
- To convert from SI units (μg m-2) used in the present study, to units of μg ft-2 used in the US, multiply loading values expressed in μg m-2 by a factor of 0.0929.





3. RESULTS AND DISCUSSION

3.1 Wipe Data Quality

Results indicated that 23% of Cd samples, 43% of Pb samples, and 3.6% of yttrium (Y) samples were below the respective LODs. The median of the field blanks for each element are reported in Table 1.

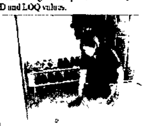
Table 1. LOD and LOQ for Cd, Pb, and Y are based on 37 analytical batches (3 procedural wipe blanks per batch). Percent recovery (mean ± sd) was calculated from 66 CRMs (NIST 2583, 2584, & 2711) (2 CRMs per batch). Results for field wipe values (median) include all field blank wipes collected in this study (n=220)

	LOD (ng m-2)	LOQ (ng m-2)	Recovery (%)	Field Wipe (ng m-2)
Cd	125	416	88 ± 14	62.5
Pb	932	3110	93 ± 6	466
Y	9.09	30.3	N/A	4.55

The relative percent difference (RPD) between co-located duplicate samples was calculated. Note that the mean RPD in the range between LOD and LOQ is greater than the mean RPD above LOQ (Table 2). The greater variability in the lower range (between LOD and LOQ) reflects a combination of field and analytical sources of uncertainty, and points to the need to consider the more rigorous LOQ as the appropriate limit for acceptance.

Table 2. Mean Relative Percent Difference (RPD) of co-located duplicates categorized by LOD and LOQ. Total number of pairs = 220. See Table 1 for LOD and LOQ values.

	Duplicates <LOD	Between LOD and LOQ		Duplicates > LOQ	
	No. of pairs	No. of pairs	Mean RPD	No. of pairs	Mean RPD
Cd	53	103	51.3	64	39.4
Pb	100	40	71.8	80	41.2



3.2 Pb and Cd Loadings

Threshold values of 125 μg m-2 (12 μg ft-2) for Pb and 4.4 μg m-2 (0.4 μg ft-2) for Cd were identified based on breakpoints in the data on the respective normality (Q-Q) plots (Figures 1 & 2). Potential sources in the homes above the thresholds for Pb (8 homes) and Cd (9 homes) include recent renovations (old paint and furnishings) and resident activities (welding, soldering, crafts, vehicle bodywork)

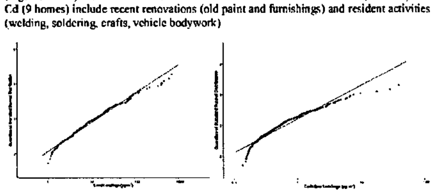


Figure 1 and 2. Normality Plot (Q-Q plot) for log transformed Pb and Cd loadings occurring in a subset of homes having at least one wipe above the LOD (n=186, n=211). Homes are represented by the wipe with the highest loading, regardless of the room from which it was taken. The line through the data represents a lognormal distribution. Eight homes are above the breakpoint in the dataset which occurs at 125 μg m-2 for Pb, and nine homes are above the breakpoint at 4.4 μg m-2 for Cd.

3.2.1 Within Home Variability

A room by room analysis was conducted with the aim of exploring potential sources of Pb and Cd (Tables 3 & 4). The entry way generally displayed higher median values for Pb and Cd suggesting outdoor sources such as track–in of dirt. The results shown in Tables 3 & 4 suggest that both indoor and outdoor sources of Pb and Cd contribute to their loadings in house dust.

Tables 3 and 4. Summary of Pb and Cd loadings (μg m-2) by room for all 222 homes sampled.

Pb	n	% < LOD	50th Percentile	95th Percentile	Max. value
Entry	208	22	3.64	87.5	619
Kitchen	218	47	1.02	21.2	165
Living room	114	49	0.922	40.9	482
Adult Bedroom	93	43	7.26	75.9	422
Child Bedroom	50	60	0.463	111	720
Play room	23	26	3.50	66.9	220

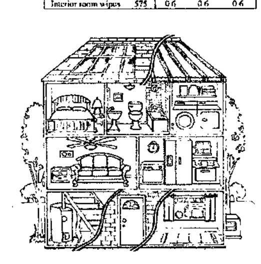
Cd	n	% < LOD	50th Percentile	95th Percentile	Max. value
Entry	208	16	0.314	3.11	8.14
Kitchen	218	25	0.207	1.34	9.84
Living room	114	20	0.239	1.20	5.66
Adult Bedroom	93	22	0.276	2.74	19.0
Child Bedroom	50	25	0.199	1.68	3.27
Play room	23	17	0.320	1.06	1.63

3.3 Elemental Associations

Spearman correlation coefficients (Table 5) were calculated to compare Cd, Pb, and Y loadings in an attempt to identify the relative contribution of outdoor and indoor sources of these metals in different areas of the home. Calabrese and Stanek (1995) determined Y to be a reliable soil tracer for use in the estimation of soil ingestion rates. The strong correlations reported in Table 5 suggest that both indoor and outdoor sources exist and thus the analysis is inconclusive as to whether indoor or outdoor sources dominate.

Table 5. Spearman rank correlation coefficients for Cd, Pb, and Y sub-divided by room. "Interior room wipes" include living rooms, kitchens, bedrooms, and play rooms.

Location	n	Cd-Y	Pb-Cd	Pb-Y
Entry wipes	208	0.6	0.7	0.7
Bedroom wipes	189	0.6	0.7	0.6
Interior room wipes	575	0.6	0.6	0.6



4. CONCLUSIONS

This research has generated the first multi-element wipe sampling database for "background" or baseline urban residential environments in Canada.

- Analysis of the maximum wipe loading per dwelling in the present study indicated that the breakpoints serve as a useful reference point for Pb (125 μg m-2) and for Cd (4.4 μg m-2).
- The empirically derived threshold for Pb is consistent with a recent American study which concluded that lead loadings less than 12 μg ft-2 (125 μg m-2) should be protective for the majority of children (Dixon et al., 2009).
- Activities conducted in each room and the products used within them contribute to the metal level of that room. Similarly, outdoor sources contribute to differences in metal loadings of homes due to track-in of outdoor dirt by residents and their pets.
- The finding of strong correlations for Y against both Pb and Cd in entry areas as well as interior areas confirms that both indoor and outdoor sources are important for these three elements.

This dataset will assist in the development of guidance about ways to reduce exposures to residential Pb and Cd that is specific to the Canadian urban environment.

5. ACKNOWLEDGMENTS

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- NSERC through Metals in the Human Environment – Strategic Network (see www.mithe-sn.org for a complete list of sponsors).
- Health Canada's Environmental and Radiation Health Science Directorate (Exposure and Biomonitoring Division), the Policy Planning and Integration Directorate (Vulnerable Populations Division), and the Safe Environments Directorate (Contaminated Sites Division).

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This study was approved by Health Canada's Research Ethics Board.

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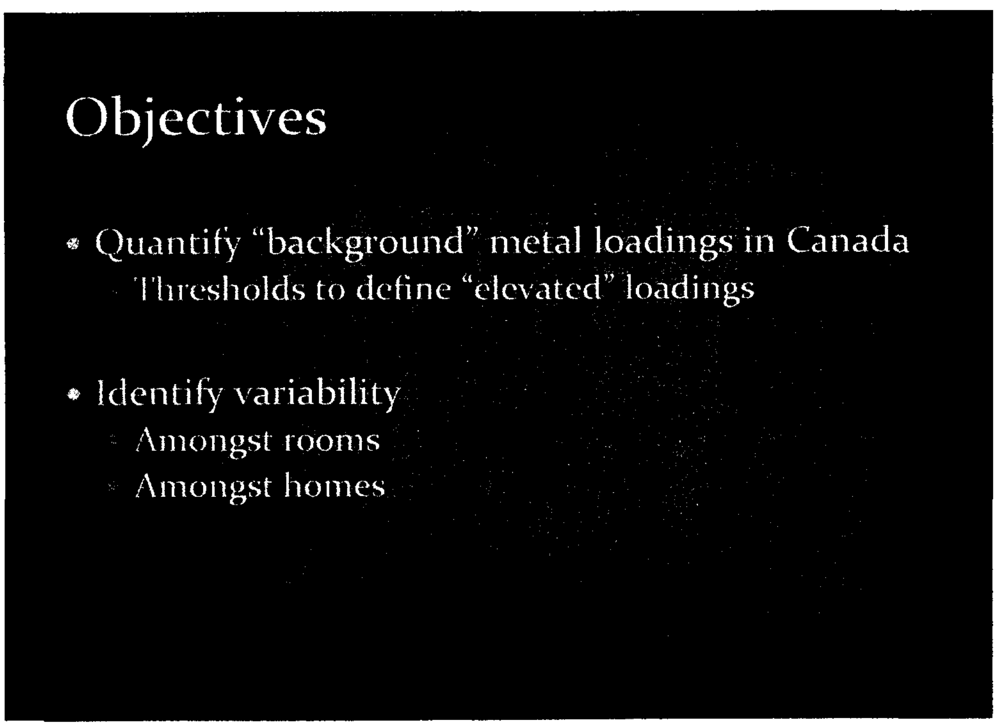
Appendix B


Wipe Sampling Methodologies to Assess Exposures to Metals in Urban Canadian Homes
Presented to: Federal Working Group on Children's Environmental Health. Nov 12, 2009, Ottawa ON.
Health Canada
Santé Canada
uOttawa
L'Université canadienne
Canada's university


Objectives
Quantify "background" metal loadings in Canada
Thresholds to define "elevated" loadings
Identify variability
Amongst rooms
Amongst homes

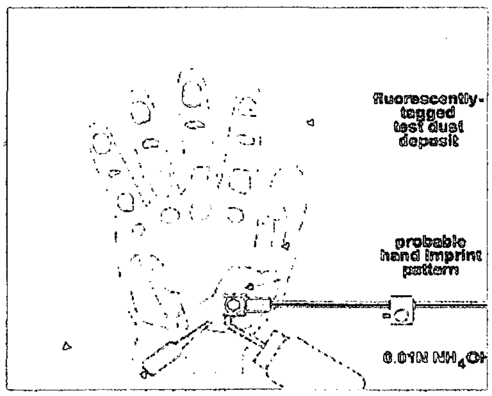
Sources of Metals in Indoor Residential Environments

- Tracking in soil and dirt
- Windblown dust
- Industrial sources
- Vehicle emissions
- Combustion processes (wood, coal, kerosene)
- Occupational ("take-home Pb")

- Consumer products
- Carpets, furnishings
- Paint, building materials
- Cooking
- Fireplaces
- Cigarette Smoke
- Crafts and Hobbies

(Rasmussen 2004)

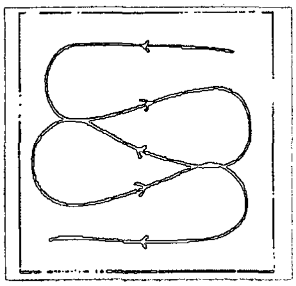
Wipes Mimic Hands



(Rodes et al. 2001)

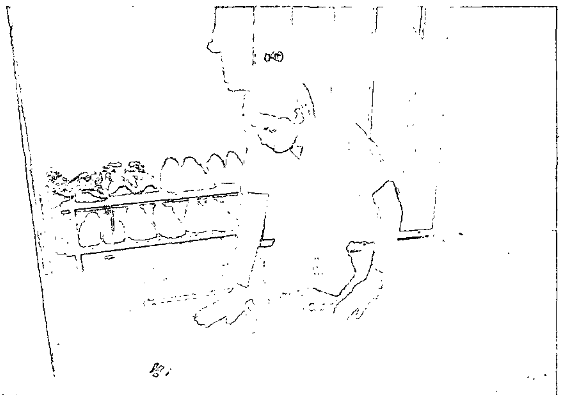
- It has been suggested that wipes more accurately mimic the motion of a child's hand
- only ~1/3 of the hand hits the surface

Sampling

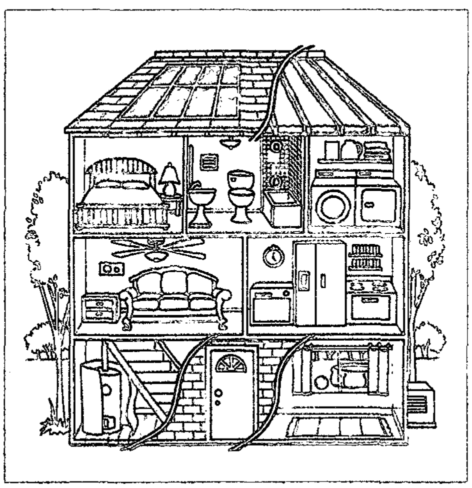


(ASTM E 1728, 2002)

- Wipe samples were taken from non-carpeted floors



Sampling Locations

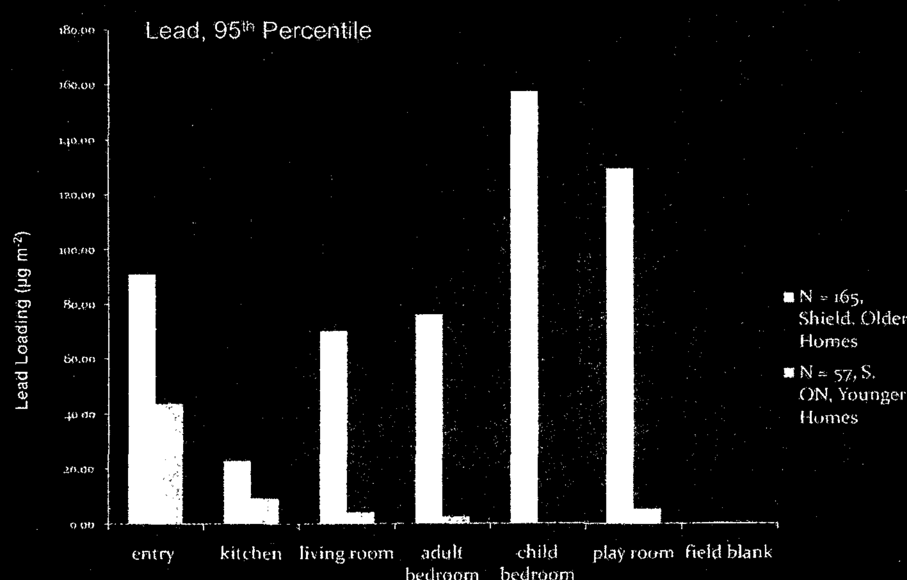


- Rooms Sampled
  - Entry
  - Kitchen
  - Living Room
  - Bedrooms
  - Play Rooms
- Place Sampled
  - Centre of floor

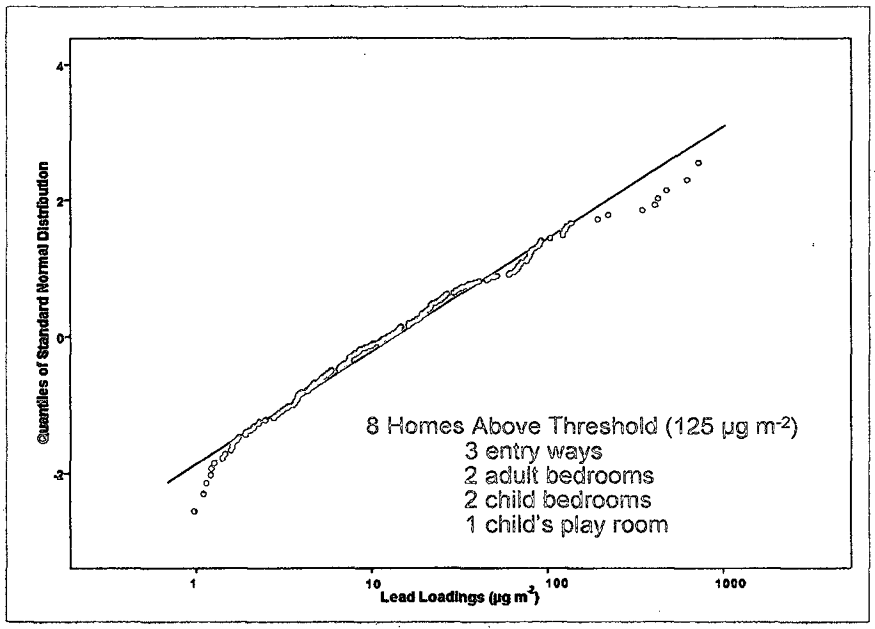
Based on advice of Wilson et al. (2006)

Results

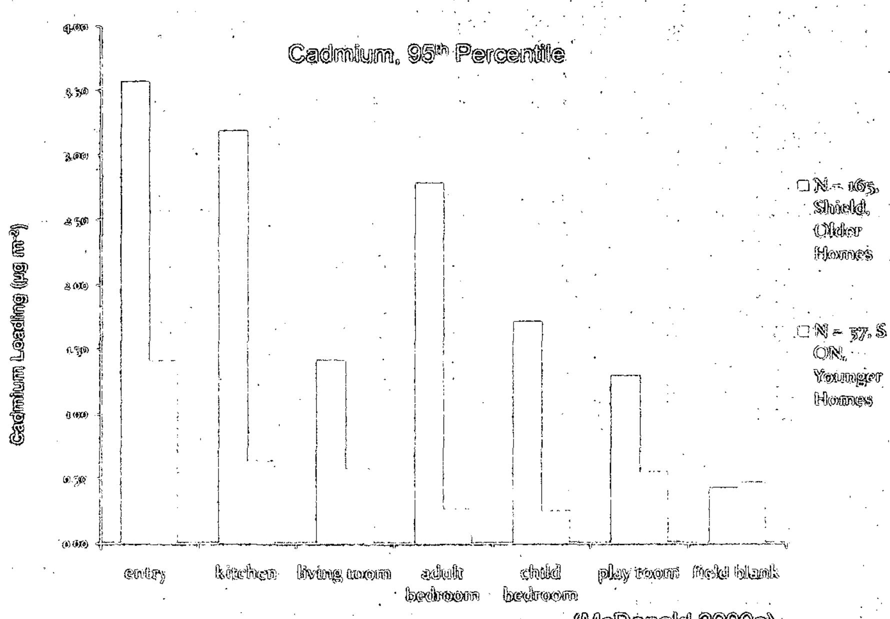
- Analysis 1:
  - Identify room – to – room variability using 95th percentiles
- Analysis 2:
  - Develop empirically derived thresholds for each metal
  - Normality (Q-Q) Plots of the max. wipe per home
  - Identify potential sources in homes with loadings above the threshold



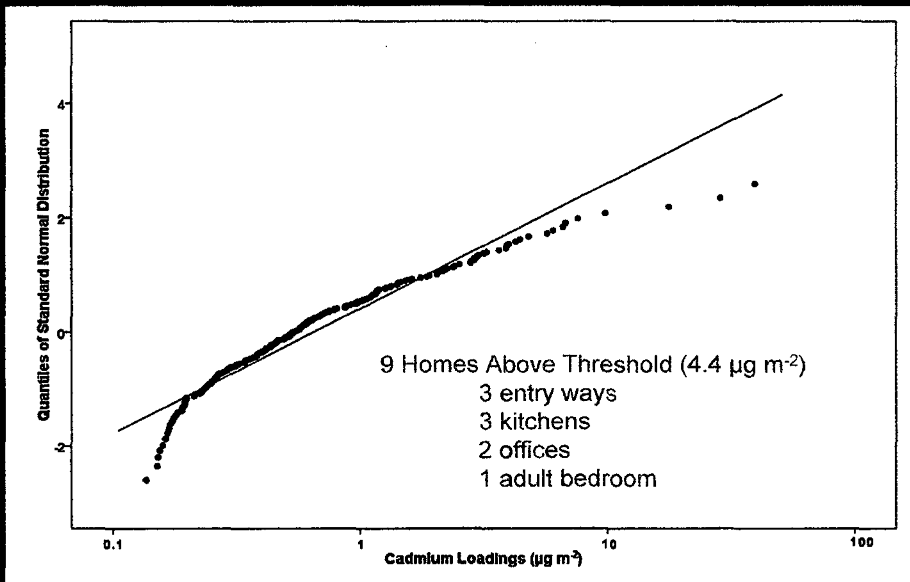
(McDonald 2009a)



(McDonald et al. 2009b)



(McDonald 2009a)



(McDonald et al. 2009b)

Conclusions

- The breakpoint in the Q-Q plot serves as a useful threshold (125 μg m^{-2} or 11.6 μg ft^{-2} for Pb)
- Consistent with Dixon et al. (2009) who concluded that a threshold of 12 μg ft^{-2} would be protective for children
- Activities conducted in each room and the products used within them contribute to the metal level of that room.
- 99% of homes in this study fell below US regulation for Pb in floor dust (USEPA, 2000). Only 3 out of 222 homes had wipe(s) exceeding the EPA regulation of 40 μg ft^{-2}.
- Similar findings as NHANES (Dixon et al. 2009).

Thanks to...

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The Canadian House Dust Study was approved by Health Canada's Research Ethics Board.

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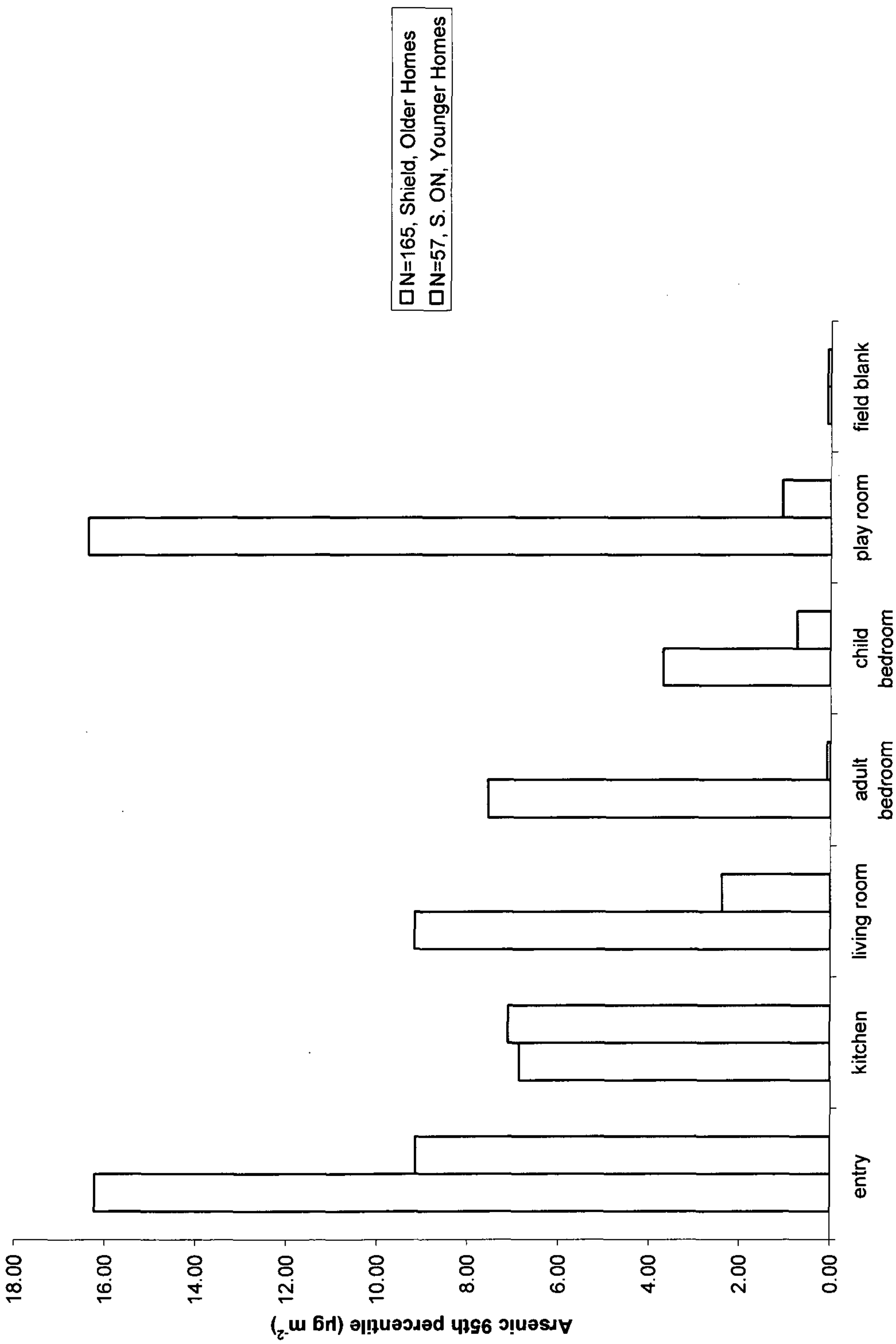
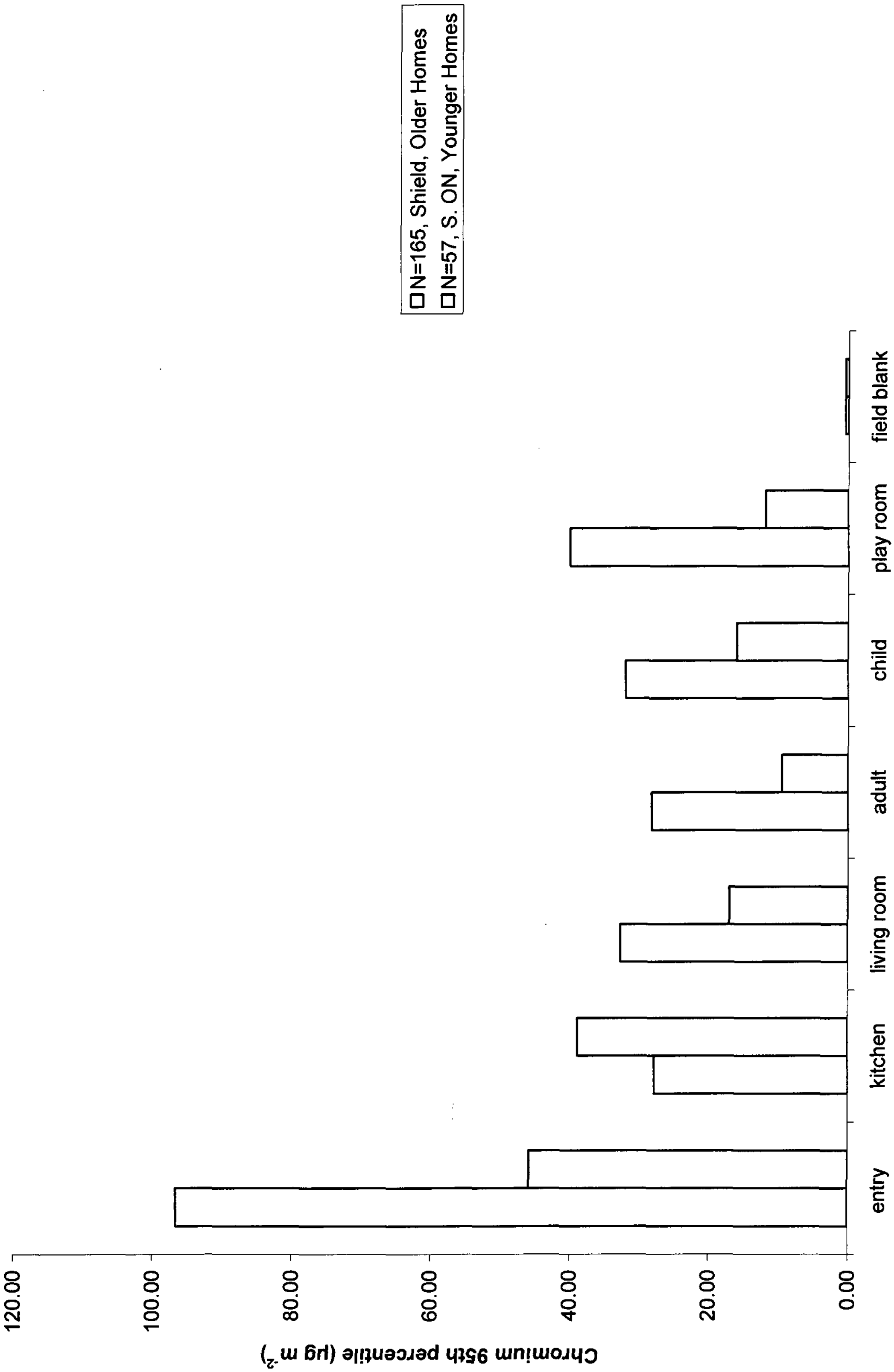
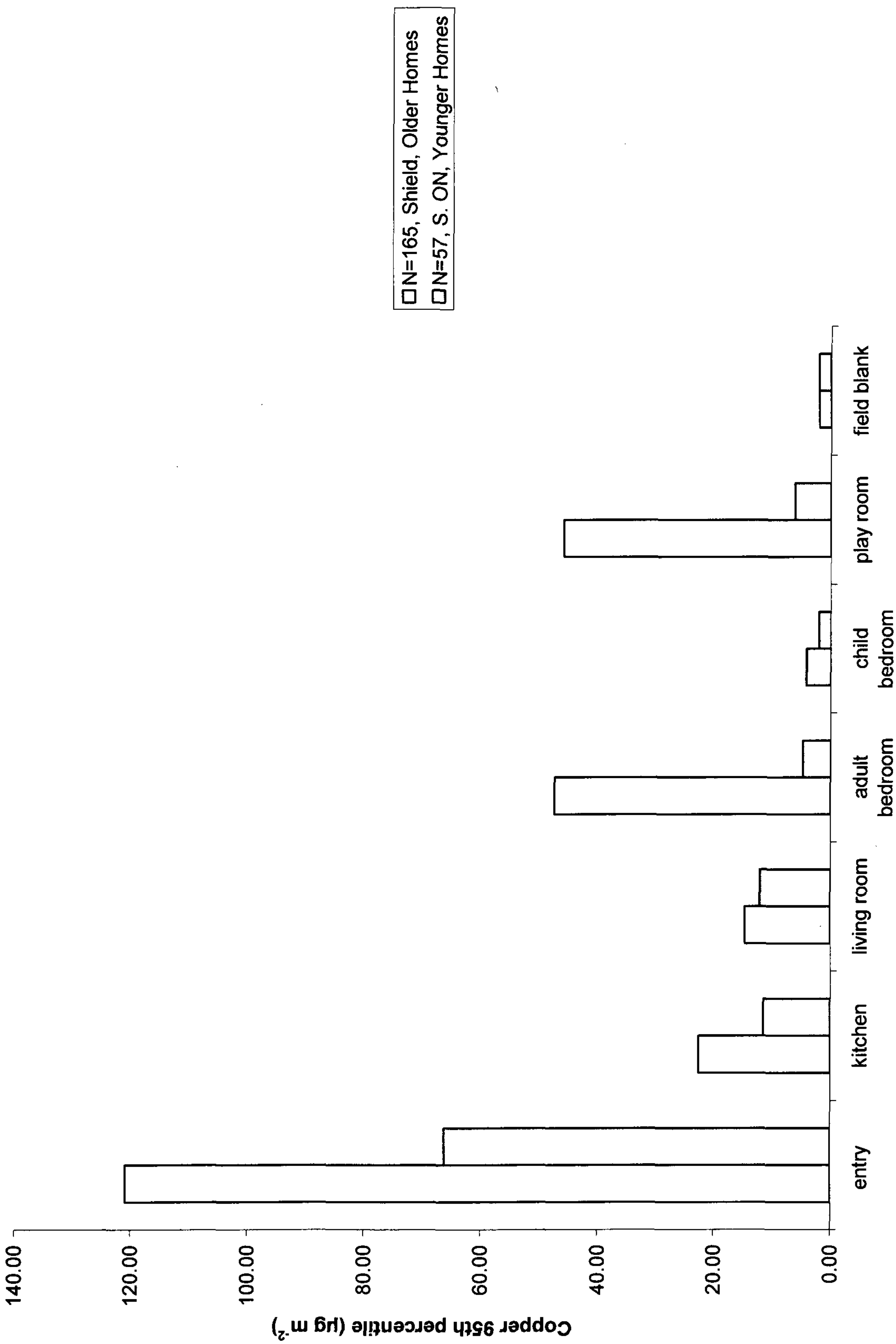
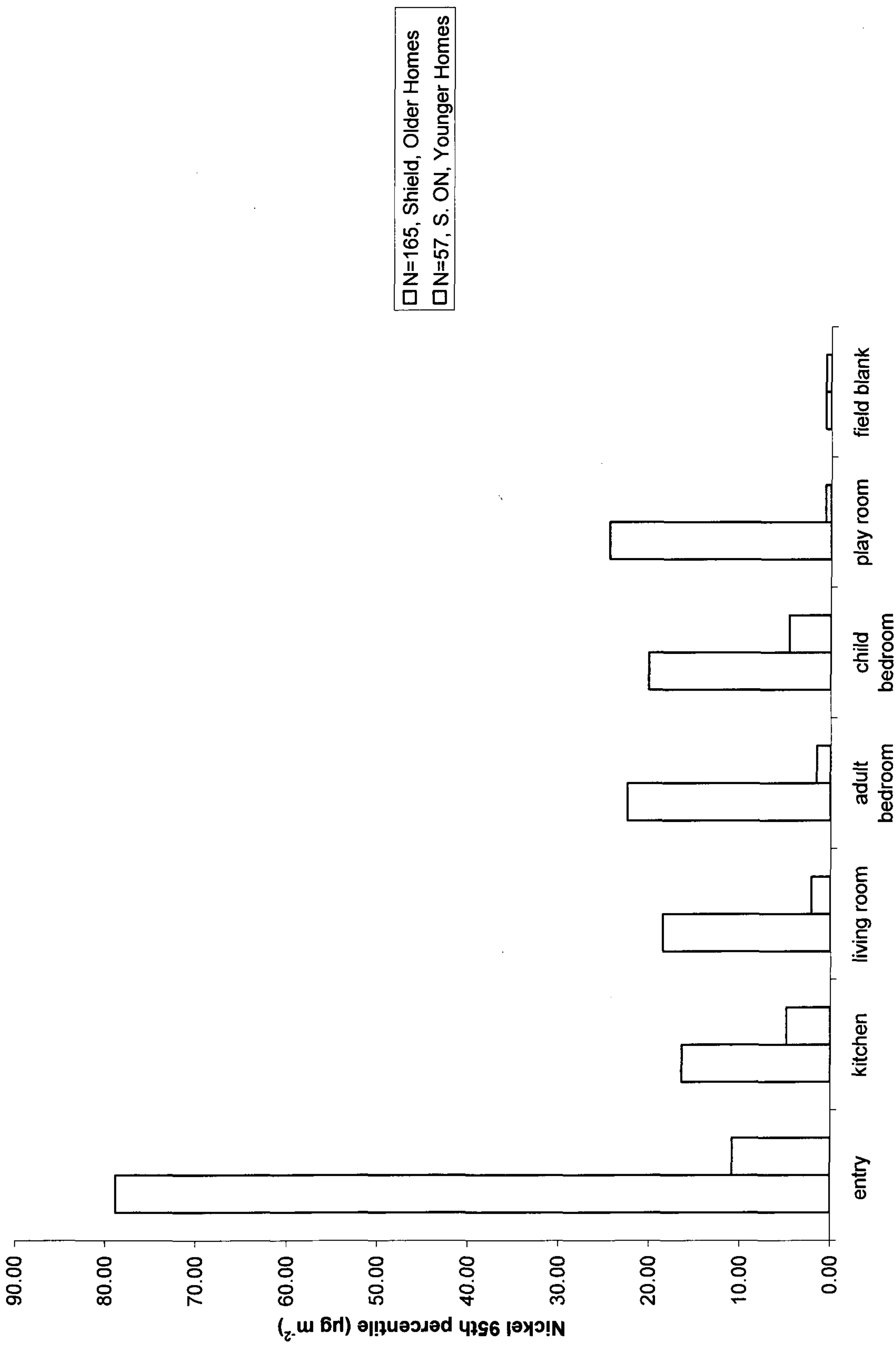
Appendix C

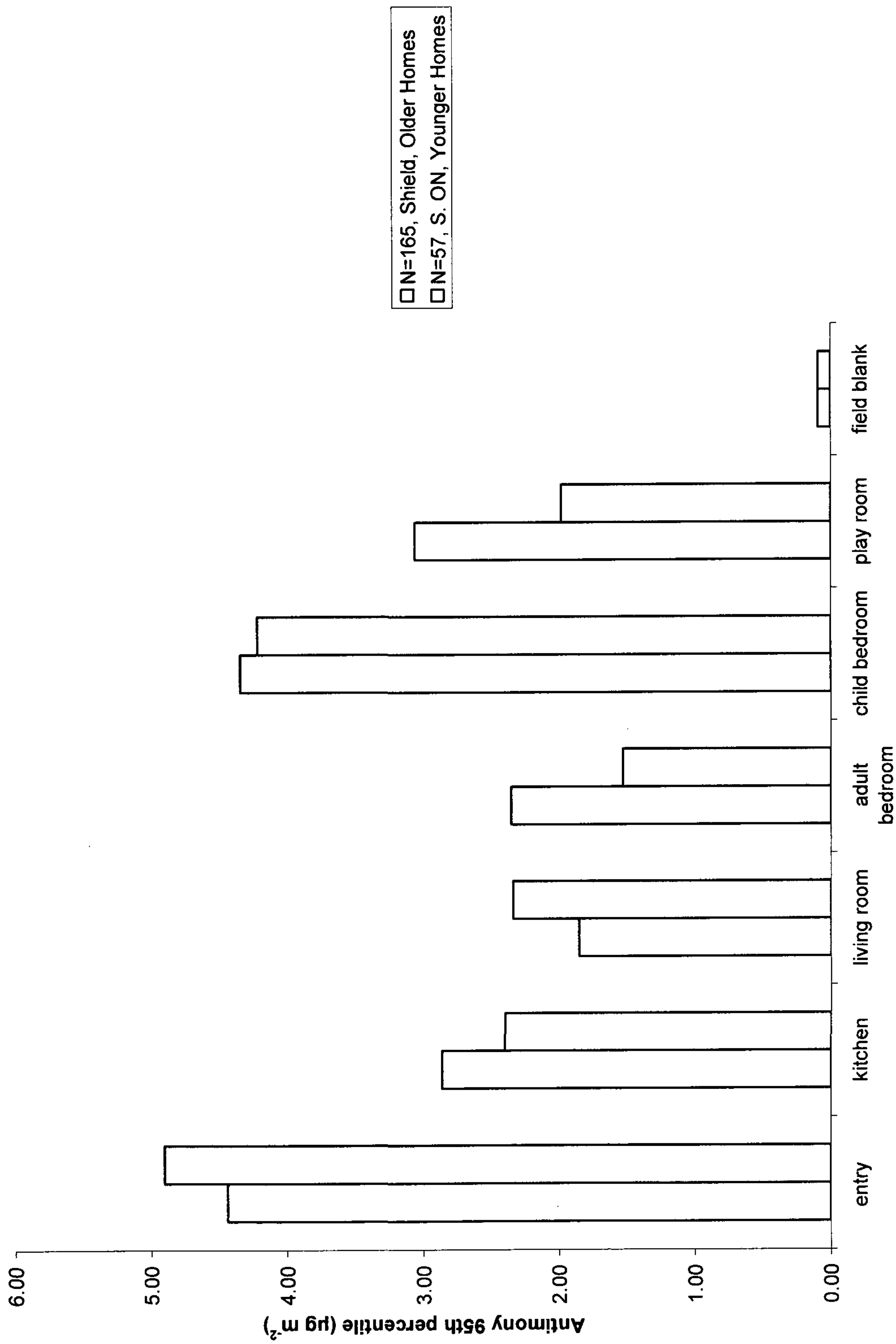


N=165, Shield, Older Homes
N=57, S. ON, Younger Homes
Arsenic 95th percentile (µg m-2)
18.00
16.00
14.00
12.00
10.00
8.00
6.00
4.00
2.00
0.00
entry
kitchen
living room
adult bedroom
child bedroom
play room
field blank


N=165, Shield, Older Homes
N=57, S. ON, Younger Homes
Chromium 95th percentile (µg m-2)
120.00
100.00
80.00
60.00
40.00
20.00
0.00
entry
kitchen
living room
adult bedroom
child bedroom
play room
field blank


Copper 95th percentile (μg m-2)
140.00
120.00
100.00
80.00
60.00
40.00
20.00
0.00
entry
kitchen
living room
adult bedroom
child bedroom
play room
field blank
N=165, Shield, Older Homes
N=57, S. ON, Younger Homes


N=165, Shield, Older Homes
N=57, S. ON, Younger Homes
Nickel 95th percentile (µg m-2)
0.00
10.00
20.00
30.00
40.00
50.00
60.00
70.00
80.00
90.00
entry
kitchen
living room
adult bedroom
child bedroom
play room
field blank


Antimony 95th percentile (µg m-2)
6.00
5.00
4.00
3.00
2.00
1.00
0.00
entry
kitchen
living room
adult bedroom
child bedroom
play room
field blank
N=165, Shield, Older Homes
N=57, S. ON, Younger Homes