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Canadian House Dust Study: Population-based concentrations, loads and loading rates of arsenic, cadmium, chromium, copper, nickel, lead, and zinc inside urban homes

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HIGHLIGHTS

- ► The Canadian House Dust Study is a nationally representative random sample.
- ► Dust was collected from 1025 urban homes from 13 cities with population > 100,000.
- ► Typical indoor dust and metal loading rates and metal concentrations are reported.
- ► Dust mass is the overriding influence on metal loadings and loading rates.
- ► This population-scale study contributes to defining the exposome.

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ABSTRACT

The Canadian House Dust Study was designed to obtain nationally representative urban house dust metal concentrations (μg g^{-1}) and metal loadings (μg m^{-2}) for arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), and zinc (Zn). Consistent sampling of active dust of known age and provenance (area sampled) also permitted the calculation of indoor loading rates (mg m^{-2} day^{-1} for dust and μg m^{-2} day^{-1} for metals) for the winter season (from 2007 to 2010) when houses are most tightly sealed. Geomean/median indoor dust loading rates in homes located more than 2 km away from industry of any kind (9.6/9.1 mg m^{-2} day^{-1}; $n = 580$) were significantly lower ($p < .001$) than geomean (median) dust loading rates in homes located within 2 km of industry (13.5/13.4 mg m^{-2} day^{-1}; $n = 421$). Proximity to industry was characterized by higher indoor metal loading rates ($p < .003$), but no difference in dust metal concentrations ($.29 \geq p \leq .97$). Comparisons of non-smokers' and smokers' homes in non-industrial zones showed higher metal loading rates ($.005 \geq p \leq .038$) in smokers' homes, but no difference in dust metal concentrations ($.15 \geq p \leq .97$). Relationships between house age and dust metal concentrations were significant for Pb, Cd and Zn ($p < .001$) but not for the other four metals ($.14 \geq p \leq .87$). All seven metals, however, displayed a significant increase in metal loading rates with house age ($p < .001$) due to the influence of higher dust loading rates in older homes ($p < .001$). Relationships between three measures of metals in house dust – concentration, load, and loading rate – in the context of house age, smoking behavior and urban setting consistently show that concentration data is a useful indicator of the presence of metal sources in the home, whereas dust mass is the overriding influence on metal loadings and loading rates.

1. Introduction

The question "What constitutes an appropriate dust sample" posed by Paustenbach et al. (1997) captures a variety of concerns about the difficulty in obtaining a standardized, representative house dust sample (Sutton et al., 1995; Butte and Heinzow, 2002), the heterogeneity of vacuum samples (Aurand et al., 1983), and the large inherent variability in dust wipe data (Caplan, 1983). Dust metal concentrations and dust metal loadings are different measures of indoor environmental metal levels: dust concentration is a measure of the amount of metal in a given quantity of dust, in units of μg g^{-1} or ppm, while loading (μg m^{-2}) is mainly a function of the quantity of dust on the surface sampled and to a lesser extent the concentration of metal in dust (Sutton et al., 1995). Both loading and concentration measures are important for understanding and

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assessing the significance of exposure to house dust (Paustenbach et al., 1997), and they provide different but complementary types of information. Loading, which is strongly influenced by housekeeping before sample collection (Sutton et al., 1995; Bell et al., 2010), is considered to be an appropriate index of potential exposure (Lioy et al., 1992; Lanphear et al., 1998; Dixon et al., 2009). Concentration information is useful for identifying residential metal sources and speciation (Sutton et al., 1995; Maclean et al., 2011; Beauchemin et al., 2011; Walker et al., 2011), and for direct comparisons of indoor dust with outdoor media such as soil and street dust (Rasmussen et al., 2001, 2008; Zota et al., 2011).

Information on everyday exposures to metals in house dust at the population scale contributes to defining the exposome, which is the measure of all the exposures of an individual in a lifetime and how those exposures relate to disease (CDC, 2012). Representative baseline information about exposures to chemicals in house dust of the general urban population has been identified as a data gap in residential risk assessments. Arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), and zinc (Zn) were selected for the present study as they commonly occur in contaminated sites in Canada (Franz Environ Inc., 2005; Rasmussen and Gardner, 2008), and urban baseline estimates for these elements are needed as a point of comparison for residential assessments on or near contaminated sites. Information about these elements is also required for calculations of population-based Estimated Daily Intakes used in guideline development.

The Canadian House Dust Study (CHDS) was designed to provide nationally representative population-based baseline survey information for typical urban Canadian homes (Rasmussen et al., 2011). In addition to reporting loading and concentration measurements, the present study also reports loading rates, which are rates of deposition expressed in units of mass per unit area per unit time (mg m^{-2} day^{-1} for dust and μg m^{-2} day^{-1} for metals). Hogervorst et al. (2007) concluded that metal loading rates should be incorporated into risk assessments, based on observed significant increases in biomarkers of Cd and Pb exposures in adults associated with increased Cd and Pb loading rates in the vicinity of smelters. There are various methods to determine dust and metal loading rates, many of which involve the collection of dust which has passively accumulated on an exposed surface area (e.g., plates, beakers, Petri dishes) over a specified period of time (Aurand et al., 1983; Edwards et al., 1998; Meyer et al., 1999; Siefert et al., 2000; Khoder et al., 2010). In the present study, trained technicians followed a vacuum sampling protocol designed to collect a consistent composite (whole-house) sample of readily accessible “fresh dust” of known age, from dry floor surfaces of known dimensions, to achieve a complete set of elemental concentration, load, and loading rate measurements.

2. Method

2.1. Sample collection

The goal of the CHDS is to obtain a population-based urban baseline estimate which is representative for Canada, not individual cities or provinces, as described previously (Rasmussen et al., 2011). Briefly, the statistical design required a stratified random sample of 1025 Canadian single family dwellings from a total of 107 dissemination areas (DAs) across 13 Canadian cities having a population greater than 100,000 (Rasmussen et al., 2011). Sampling was conducted in the winter season from January 2007 to March 2010. A composite dust sample was collected in each home using the Pullman Holt model 102 ASB-12PD vacuum sampler specified to capture 99.97% of all particles 0.3 μm and larger. The sampling method was based on the Verein Deutscher Ingenieure protocol (VDI, 2001) for sampling fresh or “active” house dust, defined as dust of known age that is readily accessible on floor surfaces of living spaces in the home. A composite sample was collected by vacuuming accessible floor surfaces in all living areas and connecting rooms, excluding potentially wet areas (kitchen, garages, workshops and unfinished sections of basements) to protect the integrity of the sample. A light touch of the vacuum nozzle was used to collect only surface dust from carpets and avoid dust of unknown age found in joints and cracks in flooring, and areas where the householder does not vacuum on a regular basis (VDI, 2001). To keep costs within the budget, vacuuming was accomplished in a single household visit rather than the two visits prescribed by the VDI protocol. Participants were requested to abstain from cleaning floor surfaces for a period of 7 days before the scheduled fresh house dust sampling, and the date of the last cleaning was documented.

2.2. Sample processing

Dust was transferred quantitatively from the vacuum bags to weighing paper, with careful avoidance of loss or contamination during processing as described previously (Rasmussen et al., 2011). The vacuum bag was cut open to allow the dust to air-dry (minimum 24 h), followed by manual removal of pet and human hair and large particles. Each sample was sieved to fine (<80 μm) and coarse (80–300 μm) size fractions using a sieve shaker (W.S. Tyler RO-Tap RX-94) and stainless steel sieves (W.S. Tyler Cat. Nos. 2451, 5209, 5205). The sieved dust samples were weighed (CP2202S Sartorius Balance; readability 0.01 g) and stored frozen in amber glass bottles with PTFE-lined caps until submission for analysis. The masses of the fine (<80 μm) and coarse (80–300 μm) dust fraction were combined to calculate the total dust load (<300 μm) collected in each home.

2.3. Elemental analysis

The analytical approaches were selected to provide a quantitative determination of total metal content in the dust samples. Instrumental Neutron Activation Analysis (INAA) is a direct solid-sample analytical technique which provides adequate detection limits for certain elements (e.g. As and Cr), but other elements require a strong acid digestion capable of putting all species of the element into solution with equal efficiency, whether the matrix is soil or dust. Total concentrations of Pb, Zn, Cu, Cd, and Ni were determined in 0.5 g subsamples of the <80 μm dust size fraction by Actlabs Inc. (Ancaster ON, Canada) using a 4-acid digestion (HF, $HClO_4$, HNO_3 and HCl) followed by either Inductively-Coupled Plasma Mass Spectrometry (ICP–MS) or Optical Emission Spectrometry (ICP OES), as appropriate for the concentration range. Arsenic was determined in 0.5 g subsamples by INAA with the exception of 62 samples which were determined using the above acid digestion/ICP method due to insufficient mass for INAA. Chromium was determined by INAA for samples ≥10 μg g^{-1} Cr, and by acid digestion/ICP–MS for samples containing <10 μg g^{-1} Cr or lacking adequate mass for INAA. Sample duplicates, in-house controls, and Certified Reference Materials (CRMs) were inserted (blind) after every 10 samples, for evaluation of reproducibility and extraction efficiency. The following National Institute of Standards & Technology (NIST) CRMs were selected to represent a wide range of total elemental concentrations in relevant matrices: indoor vacuum dust (NIST 2583 and NIST 2584), urban soil (NIST 2586, NIST 2710 and NIST 2711), and sediment (NIST 2702). All results are summarized for the individual NIST CRMs in the Supplementary information (Table S1).

2.4. Questionnaire and data analysis

During the house visit, technicians completed a short questionnaire to assist the interpretation of dust loading and composition (e.g. heating practices, dates of construction and renovation activity, proximity to industry, hobbies, number of residents and pets). Out of 1025 homes sampled, there were 3 cases where the dimensions