Environment International xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Use of Monte Carlo analysis in a risk-based prioritization of toxic constituents in house dust

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A R T I C L E I N F O

Keywords:
House dust
Risk assessment
Phthalates
Flame retardants
Children

A B S T R A C T

Many chemicals have been detected in house dust with exposures to the general public and particularly young children of potential health concern. House dust is also an indicator of chemicals present in consumer products and the built environment that may constitute a health risk. The current analysis compiles a database of recent house dust concentrations from the United States and Canada, focusing upon semi-volatile constituents. Seven constituents from the phthalate and flame retardant categories were selected for risk-based screening and prioritization: diethylhexyl phthalate (DEHP), butyl benzyl phthalate (BBzP), diisononyl phthalate (DINP), a pentabrominated diphenyl ether congener (BDE-99), hexabromocyclododecane (HBCDD), tris(1,3-dichloro-2-propyl) phosphate (TDCIPP) and tris(2-chloroethyl) phosphate (TCEP). Monte Carlo analysis was used to represent the variability in house dust concentration as well as the uncertainty in the toxicology database in the estimation of children's exposure and risk. Constituents were prioritized based upon the percentage of the distribution of risk results for cancer and non-cancer endpoints that exceeded a hazard quotient (HQ) of 1. The greatest percent HQ exceedances were for DEHP (cancer and non-cancer), BDE-99 (non-cancer) and TDCIPP (cancer). Current uses and the potential for reducing levels of these constituents in house dust are discussed. Exposure and risk for other phthalates and flame retardants in house dust may increase if they are used to substitute for these prioritized constituents. Therefore, alternative assessment and green chemistry solutions are important elements in decreasing children's exposure to chemicals of concern in the indoor environment.

1. Introduction

House dust (HD) represents a key source of children's exposure to contaminants that stem from consumer products and the built environment. The importance of HD has been shown in studies demonstrating that its levels of lead, phthalates and flame retardants track closely with the levels in children's blood or urine (Lanphear et al., 1996; Etchevers et al., 2015; Langer et al., 2014; Stapleton et al., 2012). Behavioral factors such as large percentage of time spent on the floor and frequent hand-to-mouth activity create greater exposure to HD in young children compared to older age groups (USEPA, 2011; Xue et al., 2007; Moya et al., 2004). This increases exposure to a wide range of contaminants as HD is a reservoir for semivolatile and non-volatile chemicals that are released from materials around the home (Rager et al., 2016). As such, it is not only a source of children's exposure but HD can be an indicator of the chemicals of concern to children that exist in consumer products and the built environment and thus require further source characterization (Bornehag et al., 2005; Butte and Heinzow, 2002). In fact, detection of a contaminant in HD is part of the evidence various bodies have used to prioritize chemicals as being of high concern to children (Washington State Legislature, 2008; Vermont DOH, 2015). HD concentrations can vary by region and over time reflecting local building materials, product usage and changing market conditions.

Of the variety of chemicals detected in HD, semivolatiles such as phthalates, flame retardants and perfluorinated compounds have received recent attention (Mitro et al., 2016). PCBs, pesticides, and chemicals in personal care products have also been detected (Rager et al., 2016; Knobelock et al., 2012; Ertl and Butte, 2012). The volatility of these chemicals is not high but still sufficient to create a slow release, with released chemical adsorbing onto particles and settling into HD (Xu et al., 2010). The number of different chemicals in HD and the variability in levels can make it difficult to determine which are the greatest exposure and risk to children. An evaluation of 156 chemicals from 25 chemical classes prioritized these chemicals based upon their mean HD concentrations and potency to cause chronic health effects (Bonvallot et al., 2010). This evaluation focused on HD measurements in France or across Europe where French data were not available. The

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http://dx.doi.org/10.1016/j.envint.2017.06.009
Received 1 March 2017; Received in revised form 7 June 2017; Accepted 10 June 2017

phthalate plasticizer diethylhexylphthalate (DEHP) received the highest rank score overall while the highest ranking flame retardant in house dust was pentabrominated diphenyl ether (Bonvallot et al., 2010). A recent meta-analysis of house dust data across 45 chemicals present in consumer products highlights the contributions of phthalates, phenols, flame retardants and perfluorinated compounds to indoor contaminant levels (Mitro et al., 2016). That database included studies published from 2000 forward.

The goal of the current analysis is to provide a risk-based prioritization of HD contaminants, focusing on the most recent data available from the United States and Canada. The purpose of this prioritization is to highlight contaminants for further assessment of sources and options for exposure reduction to protect children's health. Monte Carlo analysis is used to factor variability in house dust concentrations and uncertainty in the toxic potency of HD chemicals into a risk-based ranking. The current analysis focuses upon commonly detected flame retardants (N = 4) and phthalates (N = 3) that have been listed by USEPA and several states as being high concern chemicals. Variability in concentration was modeled by incorporating the distribution of HD data for a particular chemical rather than just the central estimate, while uncertainty was incorporated by including both the established potency value, if one exists, and a range of alternative values. Thus, another goal is to make maximal use of updated toxicology and epidemiology information in a risk-based ranking system. These rankings will assist in the prioritization of chemicals for more in-depth assessment especially in the area of source apportionment. Risk calculations are used solely for comparing across HD chemicals and not to estimate risks relevant to standard-setting or regulatory programs.

2. Methods

2.1. House dust database and chemical selection

A database of HD chemical concentration data was compiled based upon literature searches on Pubmed and Toxline through December 15, 2016, which included search terms “contaminants”, “chemicals”, “toxic” and target lists of chemicals and chemical groups; these terms were crossed with “dust”, “house dust”, “indoor environment”,” home”, “school” or “day care” to locate relevant studies. Target lists were modified based upon initial search results to provide a focus on those contaminants which are most frequently tested for and detected in HD. Retrieved studies were organized by date of sampling event (not publication date) and by country. For the current assessment data from the United States and Canada for the period 2008 to the present were selected to represent current and local conditions. Dust collection methods in the retrieved studies involved vacuum suction devices rather than wipe collection methods, with collection typically representing multiple portions of a building rather than one specific area (e.g., a child's bedroom).

The majority of HD studies report the median and range of detections (minimum, maximum) without providing geometric mean, arithmetic average, or percentiles of the distribution. Therefore, the central estimate for each analyte in HD was represented by the median from available studies and this was averaged across studies using sample size weight averaging so that larger studies were more influential in the calculated value.

The HD database documented chemicals that might be an important exposure source based upon frequency and magnitude of detection. An overview is presented in Fig. 1, which contains data for 24 constituents from 23 recent studies of US or Canadian homes. The arrows indicate HD chemicals which are of particular focus in this round of prioritization. These chemicals were selected based upon an initial review of chemical concentration data and the availability of toxicity information. The highest concentrations were in the phthalate group with second highest concentrations in the flame retardant group (Fig. 1). The three phthalates selected for more in depth prioritization exhibit the greatest HD concentrations and all have toxicology values available from national (USEPA) or international (European Union) bodies. Regarding flame retardants, penta-, octa-, and decabrominated diphenyl ethers have been removed from commerce in the US but still are common in HD as indicated by detections of the relevant congeners. Although BDE-209 is found at higher concentration in HD (Fig. 1), BDE-99 was selected from among PBDE congeners on the basis of greater availability of animal and human data on developmental toxicity and a lower USEPA RfD (USEPA RfD, 2008; Lopez-Espinosa et al., 2015). Other halogenated flame retardants listed by multiple authorities as being of high concern include the tris-organophosphates (TDCIPP, T-CEP, TCIPP) and HBCDD (see Fig. 1 for full chemical names). All of these have been prioritized for the current analysis with the exception of TCIPP due to gaps in the TCIPP database that are being filled by new studies at the National Toxicology Program (NTP, 2016). Other HD contaminants such as perfluorinated compounds, PCBs and pesticides were found at substantially lower concentration (Fig. 1). These and environmental phenols found in HD (Mitro et al., 2016; Rager et al., 2016) will be included in future analyses using the current methodology.

2.2. Development of distributions of house dust concentrations for Monte Carlo analysis

HD concentration data for a particular chemical were simulated with a log normal distribution in which the mean of medians across studies was used to represent the 50th percentile, the single maximum detect across all studies was assigned a value of the 99th percentile and the geometric standard deviation (GSD) was backfit to be compatible with the 50th and 99th percentiles. This procedure makes the maximum detect particularly influential. To ensure that this value was not an extreme value, the next highest detection was identified. In all cases, the maximum detect was within 4 fold of the next highest value indicating that the selected value is not an extreme outlier. For one chemical, DINP, only one study was available but the maximum value is supported by a similar maximum from a recent study from Japan (Ait Bamai et al., 2014). Confidence in the constructed distributions is increased by the fact that the GSDs resulting from this backfitting procedure were found to be within range of the actual GSDs in the few cases where this was available. For example, the modeled GSD pooled across BDE-99 studies was 1.73 (Table 1) while available GSDs were 1.47 (Stapleton et al., 2012, raw data sent from author) and 1.53 (Shoeib et al., 2012). For HBCDD the backfit GSD was 1.25 while in Shoeib et al. (2012) it was 1.05. The median and GSD values shown in Table 1 were then used to construct the remainder of the lognormal distribution for each HD chemical using the Excel log-inverse function, which provided HD concentrations associated with percentiles 1 through 100 of the lognormal distribution as described previously (Ginsberg, 2012).

2.3. Use of Monte Carlo analysis to prioritize HD chemicals

A risk-based approach was used to compare across chemicals and provide relative rankings. The risk characterization equation shown below estimates hazard quotient (HQ) as the ratio of exposure dose for young children (0–6 years of age) divided by the reference dose (non-cancer endpoints) or de minimis cancer risk dose as defined below. The exposure portion of the equation (the numerator) is based solely on a child's ingestion of chemical in house dust. While inhalation and dermal exposure may also occur, this would require a more comprehensive multi-pathway analysis of HD contaminants which was beyond the current scope. However, the ingestion pathway is expected to provide a reasonable representation of the relative importance of these semi-volatile HD contaminants given that other pathways (e.g., dermal, inhalation) will have a similar relationship to ingestion across chemicals. The one exception is TCEP which has a greater distribution into indoor