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Indoor residential exposure to semivolatile organic compounds in France

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ABSTRACT

Multiple chemicals are emitted in residential accommodation. Aggregate Daily Doses (ADD) (ng/kg-bw/d) were estimated for 32 semivolatile organic compounds (SVOCs) of different chemical families that are frequently detected in French dwellings in both air and settled dust. Daily doses were determined using steady-state models for the population, categorized into 11 age groups covering birth to age 30. Three routes of exposure were taken into account: dust ingestion, inhalation (gaseous and particulate phases) and dermal contact with the gaseous phase of air. Contamination levels were preferentially retrieved from large, nationwide representative datasets. A two-dimensional probabilistic approach was used to assess parametric uncertainty and identify the most influential factors. For children aged 2 to 3 years, ADD estimates spanned orders of magnitude, with median values ranging from 8.7 pg/kg-bw/d for 2,2′,3,4,4′-pentabromodiphenylether (BDE 85) to 1.3 µg/kg-bw/d for di-isobutyl phthalate (DiBP). Inhalation, ingestion and dermal pathway contributed at varying levels, and depending on compound, air was the dominant medium for 28 of the 32 compounds (either by inhalation or dermal contact). Indoor exposure estimate variance was mainly driven by indoor contamination variability, and secondarily by uncertainty in physical and chemical parameters. These findings lend support to the call for cumulative risk assessment of indoor SVOCs.

1. Introduction

Both consumer product use and the production of chemicals have been rising constantly since the mid-20th century, and many of these chemicals are semivolatile organic compounds (SVOCs). SVOCs include compounds from various chemical families: phthalates, bisphenols, polycyclic aromatic hydrocarbons (PAHs), organophosphorus (OPs), organochlorines (OCs), synthetic musks, polychlorinated biphenyls (PCBs) and polybromodiphenylethers (PBDEs).

The health effects of SVOCs have been assessed by numerous studies on both humans and animals; some are suspected of having reprotoxic (Casas et al., 2013; Rubin, 2011), neurotoxic (Baldi et al., 2001; Blanc-Lapierre et al., 2012; Elbaz et al., 2009; Zaganas et al., 2013) or carcinogenic effects (Armstrong et al., 2004; IARC, 2015a, 2015b).

SVOCs are emitted by volatilization from their source materials and contaminate other compartments; in some cases, they can also migrate directly from source (Sukiene et al., 2017). In the indoor environment, they are found in the gas phase, airborne particles, settled dust (Weschler and Nazaroff, 2008) and on any other available surfaces such as walls, ceiling and flooring – as well as on human skin and clothing. In addition to dietary exposure and dermal contact with consumer products, humans are continuously exposed to these chemicals through various pathways, including inhalation of indoor air (gaseous and particulate phases), ingestion of settled dust and dermal contact with indoor air and settled dust (on floor and other surfaces). Many authors have assessed indoor exposure to certain families of SVOCs (phthalates, PBDEs, etc.), taking into account one or more exposure media via oral, respiratory (and sometimes dermal) pathways (Bekö et al., 2013; Gaspar et al., 2014; Linares et al., 2010; Mitro et al., 2016; Roosens et al., 2010; Trudel et al., 2011; Wilson et al., 2003). Mitro et al. (2016) recently estimated indoor exposures based on US dust surveys and an air contamination model. Here, we seek to use measurement data to assess the exposure of a large population and estimate the associated uncertainty.

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The objective of this study was to estimate the indoor exposure of people of various age groups, to 32 SVOCs from different chemical families frequently detected in French dwellings (Blanchard et al., 2014; Mandin et al., 2014, 2016). Three routes of exposure were taken into account: dust ingestion, inhalation of air (gaseous and particulate phases) and dermal contact with the gaseous phase of air. Contamination levels were preferentially retrieved from large, representative datasets. A two-dimensional probabilistic approach was used to assess the uncertainty associated with the different parameters, and identify the most impacting factors.

2. Methods

2.1. Target population

To address exposure across a broad section of the population, we estimated exposures for 11 age groups from birth to age 30 (as an example of an adult), following the U.S. Environmental Protection Agency (U.S. EPA, 2005) recommendations as to which age groups should be considered within a health risk assessment.

2.2. Compounds selection

32 SVOCs were selected on the basis of their health interest (Bonvallot et al., 2010), and because they were detected in both the air and the settled dust of French dwellings (Blanchard et al., 2014; Mandin et al., 2014, 2016).

2.3. Exposure model

The Aggregate Daily Doses (ADD) (ng/kg-bw/d) were assessed by summing internal (uptake) daily doses from dust ingestion ($DD_{ing\text{-}dust}$), inhalation of air (both gaseous and particulate phases) ($DD_{inh\text{-}air}$) and dermal contact with gas phase ($DD_{derm\text{-}gas}$).

Very few studies included the dermal exposure to dust pathway when assessing aggregate exposures to SVOCs. Trudel et al. (2011) studied dermal exposure to 8 PBDEs in dust. Even though they overestimated this pathway using in vitro experimental data with acetone as carrier vehicle (Roper et al., 2006), they found the contribution of dermal exposure to dust to be consistently < 20%, even for the most contaminated region, and for every age group (below 1 year to 65 years of age). Bekö et al. (2013) estimated indoor exposure to five phthalates and found a very low (< 1%) contribution of dermal exposure to dust, in comparison to other pathways. Since this pathway is typically found to be minor and required uptake parameters are ill-suited to dust exposure even when available, dermal exposure to dust was not addressed in this work.

2.4. Equations for exposure dose estimation

Daily Doses (DD) can be estimated in steady-state conditions using the following equations. These were adapted from relationships developed by Bekö et al. (2013) and Weschler and Nazaroff (2012, 2014).

Ingestion of settled dust:

$$DD_{ing\text{-}dust} = \frac{C_{dust} \times DI \times f_{oral} \times f_{dust} \times t}{BW} \quad (1)$$

where C_{dust} is the SVOC concentration in settled dust (ng/g), DI is the amount of dust ingested by an individual per day (g/d), f_{oral} is the oral bioavailability of the SVOC (−), f_{dust} is the bioaccessibility of the SVOC from the dust (−), t is the fraction of time spent in dwellings (−), BW is the body weight (kg), and $DD_{ing\text{-}dust}$ is expressed in ng/kg-bw/d.

Inhalation of indoor air:

$$DD_{inh\text{-}air} = \frac{(C_{part} + C_{gas}) \times IR \times f_{pulm} \times t}{BW} \quad (2)$$

where C_{part} is the SVOC particulate phase concentration (ng/m^3), C_{gas} is the SVOC gas phase concentration (ng/m^3), IR is the inhalation rate for an individual per day (m^3/d), f_{pulm} is the pulmonary bioavailability of the SVOC (−), t is the fraction of time spent in dwellings (−), BW is the body weight (kg), and $DD_{inh\text{-}air}$ is expressed in ng/kg-bw/d.

Dermal contact with the gas phase:

$$DD_{derm\text{-}gas} = \frac{C_{gas} \times k_{p\text{-}g} \times BSA \times t}{BW} \quad (3)$$

where C_{gas} is the SVOC gas phase concentration (ng/m^3), $k_{p\text{-}g}$ is the SVOC transdermal permeability coefficient (m/h), BSA is the body surface area (m^2), t is the daily duration exposure (h/d), BW is the body weight (kg), and $DD_{derm\text{-}gas}$ is expressed in ng/kg-bw/d. The steady-state model adapted by Weschler and Nazaroff (2012, 2014) used to estimate $k_{p\text{-}g}$ is described in more detail in Supplemental Material (see S1). This requires use of the SVOC octanol/water partition coefficients (log (K_{ow})), Henry's law constants (H) and coefficients describing the external transport of a gaseous SVOC from the bulk indoor air to the boundary layer adjacent to the skin (γ_d).

The ADD for a single SVOC for an individual was then calculated by summing the previous doses according to the following equation, and expressed in ng/kg-bw/d:

$$ADD = DD_{ing\text{-}dust} + DD_{inh\text{-}air} + DD_{derm\text{-}gas} \quad (4)$$

2.5. Parameter estimation for exposure model

Parameter distributions were constructed or retrieved from the literature as detailed below. Some of these parameters will be the same for all SVOCs (γ_d, BW, BSA, IR, DI and t) while others will vary from one compound to another (f_{oral}, f_{dust}, f_{pulm}, log (K_{ow}), H, C_{dust}, C_{part} and C_{gas}).

2.5.1. Physical and chemical parameters

For each SVOC, measured or estimated values of log (K_{ow}) and H at 25 °C were retrieved from: online databases - Hazardous Substances Data Bank (HSBD) and ChemIDplus (http://toxnet.nlm.nih.gov/), Chemspider (http://www.chemspider.com/), and Chemicalize (http://www.chemicalize.org/); toxicological and environmental data sheets from the French National Competence Center for Industrial Safety and Environmental Protection (INERIS) (http://www.ineris.fr/substances/fr/page/21); online calculators - Chemexper (https://www.chemexper.com/) and ACD/Labs (http://www.acdlabs.com/); EPI Suite software (U.S. EPA, 2013, v4.1) and the Handbook of Physical-Chemical Properties and Environment Fate for Organic Chemicals (Mackay et al., 2010a, 2010b, 2010c, 2010d). Particular attention was paid to avoiding duplicates (EPI Suite software, HSBD and ChemIDplus often used the same sources for these parameters). Furthermore, only values at the reference temperature of 25 °C were selected, in order to obtain comparable data between compounds and estimate DD at a constant temperature. At least two values were available for each SVOC. Where at least 15 values for log (K_{ow}) and H were available, distributions were fitted. Otherwise, we used either triangular distributions having at least three values (minimum, average and maximum), or uniform distributions for two retrieved values. See Table S1 for corresponding distributions and input parameters for each SVOC.

2.5.2. Contamination data

Contamination data were provided from measurements taken in recent French housing surveys. Concentration levels in settled dust collected from vacuum cleaner bags were retrieved from a national survey covering the 3.6 million French dwellings that were home to at least one child aged 6 months to 6 years in 2008–2009, using 145 samples (Mandin et al., 2014). Concentration levels in airborne Particulate Matter (PM) of 10 μm in diameter were retrieved from a national survey covering the 24.7 million French main residences, using 285