Environment International 94 (2016) 508–518

Contents lists available at ScienceDirect

Environment International

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

Coupled near-field and far-field exposure assessment framework for chemicals in consumer products

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ARTICLE INFO

Article history:
Received 15 April 2016
Received in revised form 2 June 2016
Accepted 8 June 2016
Available online 16 June 2016

Keywords:
Near-field consumer exposure
Multimedia multi-pathway framework
Product intake fraction
Life cycle impact assessment
Chemical alternatives assessment
High-throughput risk screening

ABSTRACT

Humans can be exposed to chemicals in consumer products through product use and environmental emissions over the product life cycle. Exposure pathways are often complex, where chemicals can transfer directly from products to humans during use or exchange between various indoor and outdoor compartments until sub-fractions reach humans. To consistently evaluate exposure pathways along product life cycles, a flexible mass balance-based assessment framework is presented structuring multimedia chemical transfers in a matrix of direct inter-compartmental transfer fractions. By matrix inversion, we quantify cumulative multimedia transfer fractions and exposure pathway-specific product intake fractions defined as chemical mass taken in by humans per unit mass of chemical in a product. Combining product intake fractions with chemical mass in the product yields intake estimates for use in life cycle impact assessment and chemical alternatives assessment, or daily intake doses for use in risk-based assessment and high-throughput screening. Two illustrative examples of chemicals used in personal care products and flooring materials demonstrate how this matrix-based framework offers a consistent and efficient way to rapidly compare exposure pathways for adult and child users and for the general population. This framework constitutes a user-friendly approach to develop, compare and interpret multiple human exposure scenarios in a coupled system of near-field ('user' environment), far-field and human intake compartments, and helps understand the contribution of individual pathways to overall human exposure in various product application contexts to inform decisions in different science-policy fields for which exposure quantification is relevant.

1. Introduction

Chemicals in products have the potential to expose consumers through product use and the general population through emissions to the environment (Bergfeld et al., 2005; Fantke and Jolliet, 2016; Geueke et al., 2014; Molander et al., 2012; Nazaroff and Weschler, 2004). Quantitatively evaluating near-field consumer and far-field population exposures to product-related chemicals is relevant for various science-policy fields including life cycle impact assessment, LCIA (Ernstoff et al., 2016), chemical alternatives assessment, CAA (Lavoie et al., 2010), risk-based assessments and high-throughput screening, HTS (Dionisio et al., 2015; ECHA, 2012; Shin et al., 2015). LCIA aims at considering exposures along all product life cycle stages and accounts for exposures mediated through environmental emissions based on interconnected compartments, but currently mostly fails to consider exposures to chemicals from product use (Jolliet et al., 2015). In risk-oriented assessments, multiple exposure pathways can be addressed, but typically in a disconnected way based on different modeling systems and levels of detail (van Leeuwen and Vermeire, 2007). For example, recent HTS approaches include multi-pathway consumer exposure, but pathways like inhalation and dermal permeation are either decoupled using two different probabilistic modeling strategies without a unifying mass balance equation (Isaacs et al., 2014), or consumer use scenarios are not combined with post-use environmental emissions (Delmaar et al., 2013). In CAA, exposure to product-related chemical emissions is currently mostly not considered or restricted to qualitative evaluations of consumer exposure (Jacobs et al., 2016).

Comprehensively accounting for the all relevant transfers and exposure pathways, which build on different underlying models, is therefore an unresolved problem across science-policy fields. Exposure pathways can involve multiple indoor and outdoor transfers before resulting in human consumer or population exposure. Near-field consumer exposure thereby refers to chemical intakes by humans using a considered product or in the vicinity of product use, and far-field population exposure refers to aggregated intakes by humans (product users and non-users) via environmental emissions. Pathways within the vicinity of product use, such as inhalation of chemicals released from products

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http://dx.doi.org/10.1016/j.envint.2016.06.010

indoors, skin permeation of chemicals in dermally applied products, and ingestion of chemicals in food and beverages often dominate exposure (Fantke et al., 2011; Wambaugh et al., 2013). Nevertheless, exposure following chemical emissions to the environment can in certain cases be of comparable magnitude depending on the product-chemical combination (Ampleman et al., 2015; Jones-Otazo et al., 2005). Considering and comparing multiple exposure pathways is therefore critical to capture overall exposure to chemicals in consumer products and to identify dominating exposure pathways. The diverse nature of exposures to chemicals from products in the vicinity of consumers, however, poses a challenge for consistently integrating models for multiple near-field pathways in a compatible way with models for far-field pathways mediated through environmental emissions.

To rigorously address this problem, a framework is needed that fully and consistently integrates different models for consumer and population exposures. To accommodate future developments and the needs of specific assessments it is important that such a framework allows for flexibility, while at the same time maintaining mass balance and physicochemical principles across inter-compartment transfers and exposure pathways capturing chemical fate indoors and outdoors. Flexibility is important with respect to data input and model selection to align with specific assessment goals, e.g. worst-case or average product use scenarios. Finally, to allow for consistent comparison across exposure pathways, chemicals, and products, such a framework should be based on comparative exposure metrics like the recently proposed product intake fraction (Jolliet et al., 2015), defined as chemical mass taken in by humans – via all relevant exposure pathways during and after product use – per unit mass of chemical in the product.

In response to these needs, we propose a novel multimedia modeling framework that couples pathways accounting for human exposures to chemicals in consumer products. First, the overall framework is defined, structuring exposure pathways as interactions between chemicals, products, environmental compartments, and humans, in order to follow the pathway from the mass of chemical used in a product to ultimate human intake. Second, after quantifying the mass of chemical used, the mathematical framework is presented to calculate exposure pathway-specific product intake fractions (PiFs) and intakes based on a multi-compartmental transfer fractions matrix approach. Finally, the applicability of our framework is illustrated in two examples of chemicals in cosmetics and flooring materials. This framework sets up a flexible structure to consistently quantify and compare any consumer product human exposure to its chemical constituents via all relevant product use-related exposure pathways.

2. Methods

2.1. Product intake fraction framework

Building on a consistent set of terms defined in Table 1, we present a comprehensive Product Intake Fraction Framework for coupled consumer and population exposures to chemicals in consumer products (referred to as *PiF Framework*) following three main steps (Fig. 1). The near-field environment is represented by a set of *near-field compartments* in the vicinity of product users. In the PiF Framework, we first quantify the chemical mass that enters a defined *compartment of entry* (Fig. 1a; Table 1). Second, the framework captures the fate and transport processes resulting in transfers of chemicals between any near-field and far-field compartment, until finally reaching humans. The combination of various multimedia transfers expressed as *direct transfer fractions* yields *cumulative transfer fractions* or, when received by human intake compartments, cumulative *product intake fractions* describing *exposure pathways* from the compartment of entry to humans via specific

Table 1
Definition of terms used in the PiF Framework for assessing human exposure to chemicals in consumer products.

Term	Definition
Compartment terms	
Near-field compartment	Any ***indoor or near-consumer*** location or environment within the vicinity of the use of a considered product ('user' environment), to and from which chemical transfers occur and within which removal processes occur. Near-field compartments include indoor air, consumer products and objects themselves, and their surfaces (e.g. 'skin surface layer' for products applied on top of the skin surface, or 'article interior' for products like flooring materials).
Far-field compartment	Any location or environment that is ***distant from the use of a considered product***, to and from which chemical transfers occur and within which removal processes occur. Far-field compartments include environmental media (e.g. 'ambient air', 'freshwater' like rivers and lakes, or 'soil'), biota (e.g. agricultural crops, wild animals and plants), or technological systems (e.g. waste water treatment plants and landfills).
Human intake compartment	Any physical location in the ***interior of humans via which the chemical is first taken in***, representing a specific route of chemical intake into the human body. Human intake compartments include the 'respiratory tract' for inhalation, 'gastrointestinal (GI) tract' for ingestion, and skin 'epidermis' for dermal uptake.
Compartment of entry	Compartment into which or within which a ***chemical is first applied or used*** (e.g. 'article interior' for a chemical embedded in flooring material or 'skin surface layer' for chemicals in dermally applied cosmetics) and from which exposure to chemicals in consumer products originates following different pathways. The compartment of entry is specific to a product use scenario.
Transfer source compartment	Any compartment, from which a chemical mass transfer occurs.
Transfer receiving compartment	Any compartment, to which a chemical mass transfer occurs.
Inter-compartment transfer terms	
Direct transfer fraction	Chemical mass fraction in any compartment that is ***passing a boundary to any adjacent or nested compartment*** via advective, diffusive, or chemical processes, such as deposition, volatilization or dermal uptake. Direct transfer fractions can be derived from different types of models (e.g. ratios of rate constants). For each transfer source compartment, all direct transfer fractions plus the fractions lost (e.g. by degradation) that are incorporated in the underlying transfer fractions models plus the fraction left in the product always sum up to unity (100%). All models used to determine direct transfer fractions need to build on ***mass balance principles*** to ensure overall consistency.
Cumulative transfer fraction	Overall chemical mass fraction originating from a certain source compartment that ***eventually reaches*** any ***receiving near-field, far-field or human intake compartment*** as a combination of all involved multimedia transfers.
Human exposure terms	
Product intake fraction (PiF)	Chemical mass within a product that is eventually taken in by humans per unit of chemical mass in that product (Jolliet et al., 2015), determined here as the cumulative transfer fraction from a compartment of entry that ***eventually reaches a certain human intake compartment*** of the considered exposed users or population group.
Exposure pathway	The course a chemical takes from its source to the person(s) being contacted (US-EPA, 1992), determined here as a ***sequence of inter-compartmental chemical transfers*** originating from a product and ending with human intake. An example exposure pathway is a chemical originally encapsulated in a solid object defined as compartment of entry 'article interior', which is then emitted to 'indoor air' and subsequently inhaled by product users, or further transferred to 'ambient air' and inhaled by the general population.
Exposure route (or route of exposure)	A particular ***means of intake*** for a chemical into the human body (US-EPA, 1992), i.e. inhalation via breathing; ingestion via intentional dietary consumption or inadvertent non-dietary intake; dermal contact between skin and external surfaces, air or products; or injection (mainly in the medical domain, e.g. vaccination).