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A semi-probabilistic modelling approach for the estimation of dietary exposure to phthalates in the Belgian adult population

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ABSTRACT

In this study, a semi-probabilistic modelling approach was applied for the estimation of the long-term human dietary exposure to phthalates – one of world's most used families of plasticisers. Four phthalate compounds were considered: diethyl phthalate (DEP), di-*n*-butyl phthalate (DnBP), benzylbutyl phthalate (BBP) and di(2-ethylhexyl) phthalate (DEHP). Intake estimates were calculated for the Belgian adult population and several subgroups of this population for two considered scenarios using an extended version of the EN-forc model. The highest intake rates were found for DEHP, followed by DnBP, BBP and DEP. In the Belgian adult population, men and young adults generally had the highest dietary phthalate intake estimates. Nevertheless, predicted dietary intake rates for all four investigated phthalates were far below the corresponding tolerable daily intake (TDI) values (i.e. P99 intake values were 6.4% of the TDI at most), which is reassuring because adults are also exposed to phthalates via other contamination pathways (e.g. dust ingestion and inhalation). The food groups contributing most to the dietary exposure were grains and grain-based products for DEP, milk and dairy products for DnBP, meat and meat products or grains and grain-based products (depending on the scenario) for BBP and meat and meat products for DEHP. Comparison of the predicted intake results based on modelled phthalate concentrations in food products with intake estimates from other surveys (mostly based on measured concentrations) showed that the extended version of the EN-forc model is a suitable semi-probabilistic tool for the estimation and evaluation of the long-term dietary intake of phthalates in humans.

1. Introduction

Phthalates (dialkyl or alkyl aryl esters of *orhto*-phthalic acid) are one of world's most used families of plasticisers. Although mainly used to soften plastic polymers, such as polyvinyl chloride, some phthalates – especially those with a short alkyl chain length – can also be present in printing inks, lacquers, solvents, personal care products, pharmaceuticals and so on (Cao, 2010; Wormuth et al., 2006). Phthalates and their metabolites have been reported to cause detrimental effects on human health. For example, exposure to benzylbutyl phthalate (BBP) and di(2-ethylhexyl) phthalate (DEHP) is associated with increased incidences of asthma and allergies in children (Bornehag et al., 2004; Jaakkola and Knight, 2008). Phthalates are also suspected to disrupt the human endocrine system. Diethyl phthalate (DEP), di-*n*-butyl phthalate (DnBP), BBP and DEHP are even marked on the European priority list of chemicals with potential endocrine disrupting activities as Category 1 substances, meaning that evidence has been found of endocrine disrupting activity in at least one animal species, using intact animals (European Commission, 2014).

Human exposure to phthalates can occur via ingestion, inhalation, medical intravenous interventions or via dermal contact (Schettler, 2006). For most phthalates, especially DEHP, food ingestion is the most important exposure pathway (Clark et al., 2011; Fromme et al., 2007; Schettler, 2006; Wormuth et al., 2006). With respect to Belgium, dietary exposure to phthalates has already been studied in the Belgian PHTAL project for preschool children and adults (Sioen et al., 2012). In this project, exposure was calculated probabilistically by combining consumption rates of two Belgian food consumption surveys with ranges of phthalate levels measured in 572 different food products sold on the Belgian market (Fierens et al., 2012a, 2012c). Also the effect of home-cooking was considered in this dietary intake study. Analysing all the food items was laborious work, due to the complex methodology of sample handling and analysis in order to avoid

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external contamination (Fierens et al., 2012a, 2012c). Bearing this in mind, the objective of the current study is to investigate whether modelled phthalate levels in food products could be used instead for the assessment of dietary exposure to phthalates in the Belgian population.

To adequately model phthalate levels in food products, all possible contamination pathways should be considered. Phthalates may enter the food chain as a result of environmental transfer as well as via the migration from contact materials used during cultivation, production, storage or even during cooking at home (Cao, 2010; Dickson-Spillmann et al., 2009; Fierens et al., 2012c). With respect to environmental transfer modelling, various models have been developed and demonstrated for phthalates in the past (e.g., Cousins and Mackay, 2003; Fierens et al., accepted for publication; Müller et al., 2003). There are also existing various models that are able to predict the migration of phthalates from food contact materials into foods or food simulants (Oldring et al., 2014; Poças et al., 2008, 2010). However, to the authors' knowledge, there are currently no models available that consider both pathways simultaneously for the prediction of contaminant concentration levels in food products. Another thing that should be considered during modelling is the effect of processing (e.g., home-cooking) on phthalate levels in food products, since differences in phthalate intake values were noticed between intake assessment scenarios that took into account food preparation and those in which this was not considered (Sioen et al., 2012). A few years ago, a mechanistic model has already been developed for the prediction of bacterial cross-contamination during food preparation in the domestic kitchen (Mylius et al., 2007), but – to the authors' knowledge – a similar type of model is currently lacking for chemical food contaminants like phthalates.

To realise the objective of this study (i.e. to estimate human dietary exposure to phthalates by using modelled concentrations in foods), the ENvironmental Food transfer model for ORganic Contaminants (EN-forc) is used to obtain predicted levels of phthalates in Belgian food products. EN-forc is a model that predicts the occurrence of organic contaminants in environmental and agricultural media starting from observed concentrations in air, sludge, manure and concentrate and from physic-chemical property data (Fierens et al., accepted for publication). Since the EN-forc model only predicts phthalate levels in agricultural products and only as a result of environmental transfer, the modelling framework of the EN-forc model is extended by including the effect of packaging, processing and import on phthalate concentrations in foods. As a case study, the long-term dietary intake of four phthalates – DEP, DnBP, BBP and DEHP – in the Belgian adult population is estimated using a semi-probabilistic modelling approach. In addition to the predicted intake distributions of the studied phthalates, the intake estimates are evaluated against exposure limit values and the contribution of 20 different food groups to the total dietary intake of the four considered phthalates is characterised.

2. Material and methods

2.1. Framework for linking data

The framework for linking concentration and consumption data to estimate and evaluate human dietary exposure to organic contaminants with the extended version of the EN-forc model is depicted in Fig. 1. The environmental transfer part of the EN-forc model predicts concentrations of organic contaminants in environmental and agricultural media starting from observed concentrations in air, sludge, manure and concentrate and from physic-chemical property data. The modelling framework of this part has been recently described in detail by Fierens et al. (accepted for publication) and will therefore not be repeated here. The extensions to the EN-forc model are explained in the following paragraphs. The EN-forc model is implemented in MATLAB® R2011b (The MathWorks, Inc., Natick, Massachusetts, USA).

2.1.1. Food recipes

In EN-forc, food products present on the European market are formulated as food recipes, describing their composition of one or more basic agricultural, non-agricultural and/or imported food products. These recipes are all mapped to 'FoodEx2' codes. The FoodEx2 coding system was developed by the European Food Safety Authority (EFSA) as a standardised food classification and description system within the European Union (EFSA, 2011a, 2011b). EN-forc considers recipes for 1,908 different FoodEx2 codes in total, covering 20 food groups to represent the total European food market (see Table 1 for a full listing). Food recipes were gathered from various sources (cooking books, internet sources, nutrient tables and expertise of dieticians). Food recipes range from very simple, as some products are only composed of one ingredient (e.g. butter is for 100% derived from cow's milk); others are more complex like white wheat bread, which is considered to contain 60% wheat, 36% tap water, 3% yeast and 1% fat.

Lipophilic chemicals, such as phthalates, tend to concentrate in the fat matrix of a food product. For this reason, EN-forc implements fat correction factors, i.e. the ratio of the fat content of the food product and the fat content of the product's ingredient. For example, butter has a fat correction factor of 19.8 (80% fat in butter with respect to 4.04% in raw cow's milk).

Depending on the type of recipe, processing factors are taken into account for every ingredient and contaminant, since processing (e.g. washing, peeling and boiling) can alter contaminant concentrations in foods (Bayen et al., 2005; Chavarri et al., 2005; Perelló et al., 2008, 2009; Roberts et al., 2008). The EN-forc model considers 13 processing types: unprocessed, washed, peeled, cooked (not specific), baked, boiled, roasted, deep fried, shallow fried, toasted, breaded and fried, stewed and microwaved. For every food ingredient, chemical dependent processing factors are specified which will be multiplied by the contaminant concentration of an ingredient. The default value for

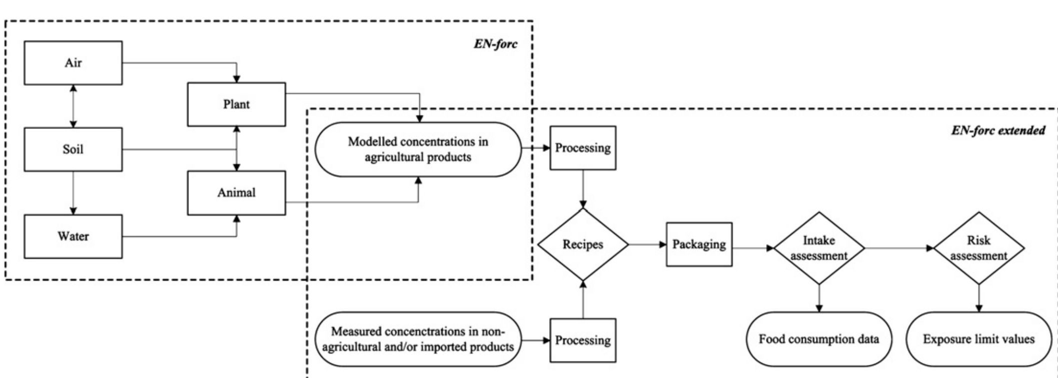

Fig. 1. Framework for linking concentration and consumption data to estimate and evaluate human dietary exposure to organic contaminants with the extended version of the EN-forc model.