Environment International 74 (2015) 249–257

Contents lists available at ScienceDirect

Environment International

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

Aggregate consumer exposure to UV filter ethylhexyl methoxycinnamate via personal care products

Eva Manová, Natalie von Goetz *, Konrad Hungerbuehler

Institute for Chemical and Bioengineering, ETH Zürich, Zürich, Switzerland

ARTICLE INFO

Article history:
Received 27 March 2014
Accepted 16 September 2014
Available online 5 November 2014

Keywords:
Ethylhexyl methoxycinnamate
UV filter
Personal care products
Aggregate exposure
Endocrine disruptor

ABSTRACT

Ultraviolet (UV) filters are substances designed to protect our skin from UV-induced damage and can be found in many categories of personal care products (PCPs). The potential endocrine-disrupting effects attributed to UV filter ethylhexyl methoxycinnamate (EHMC) are being debated. We evaluated the aggregate exposure of the Swiss–German population (N = 1196; ages ≤1–97 years) to EHMC via the use of PCPs; thus we provide the first comprehensive information about the current EHMC exposure sources and aggregate exposure levels. In our probabilistic modeling method performed at an individual level, PCP use data obtained by a postal questionnaire were linked to concentration data on EHMC gained from chemical analyses of PCPs used by the questionnaire respondents. The modeled median and 99.9th percentile of the internal aggregate exposure for the general population were 0.012 and 0.873 mg day^{-1} kg^{-1} and 0.008 and 0.122 mg day^{-1} kg^{-1} for the summer/autumn and winter/spring period, respectively. The major contributors to internal aggregate exposure were sunscreen products in summer/autumn (females: 64%; males: 85%; children aged ≤12 years 93%). In winter/spring, lip care dominated for females (30%) and sunscreen for males (38%) and children aged ≤12 years (50%). Overall, the internal aggregate exposure estimates for the studied population are shown to be below the Derived No Effect Level (DNEL) for EHMC i.e., the level of exposure above which humans should not be exposed; however, when an intense short-term exposure via sunscreen is accounted for during a sunbathing day, at the high-end percentiles (99.9th) the predicted aggregate exposure exceeds the DNEL for thyroid-disrupting effects such as for children aged ≤4 years, who might be particularly susceptible to endocrine disrupting events. It is nevertheless critical to acknowledge that quantitative data on transdermal penetration of EHMC from PCPs are currently insufficient. Since long-term effects of endocrine disruptors are not known, future studies are warranted to provide accurate quantitative data on transdermal penetration of EHMC and to determine its metabolic fate in humans.

1. Introduction

Ultraviolet (UV) filters are a class of chemicals that are increasingly added into personal care products (PCPs) (Maier et al., 2005; Séhédic et al., 2009) to protect human skin from the harmful effects of UV radiation such as immediate skin damage (sunburn), premature skin aging, and skin cancer. PCPs may also contain UV filters as UV absorbers to prevent light-induced product degradation. The worldwide UV filter production volume was estimated at about 10,000 metric tons per year (Gago-Ferrero et al., 2012). It is therefore not surprising that several UV filters are high production volume chemicals. One such chemical is ethylhexyl methoxycinnamate (EHMC) (Bachelot et al., 2012); its molecules are lipophilic (log K_{ow} 6.0; Balmer et al., 2005) and can accumulate in biota (Bachelot et al., 2012; Fent et al., 2010), with potentially adverse effects. Potential health risks to aquatic life, wildlife, and human populations associated with the possible endocrine-disrupting effects of EHMC have received wide attention not only within the scientific community, but also from the general public through media channels and non-governmental organizations (e.g., Bund für Umwelt und Naturschutz Deutschland, English: Friends of the Earth Germany, 2013; Environmental Working Group, 2014). Multiple hormonal activities of EHMC have been reported both in vitro and in vivo (Axelstad et al., 2011; Christen et al., 2011; Gomez et al., 2005; Inui et al., 2003; Klammer et al., 2007; Ozáez et al., 2013; Schlumpf et al., 2001, 2004; Seidlová-Wuttke et al., 2006), but few studies have investigated EHMC levels in human matrices (blood, urine: Janjua et al., 2004; breast milk: Hany and Nagel, 1997; Schlumpf et al., 2010), and the metabolic fate of EHMC in the human body remains unknown.

Abbreviations: PCP(s), personal care product(s); UV, ultraviolet; EHMC, ethylhexyl methoxycinnamate; SPF, sun protection factor; MCS, Monte Carlo Simulation; RCR, Risk characterization ratio; DNEL, Derived No Effect Level; US, the United States.

* Corresponding author at: Safety and Environmental Technology Group, ETH Zürich, Vladimir-Prelog-Weg 1–5/10, CH-8093 Zürich, Switzerland. Tel.: +41 44 632 0975; fax: +41 44 632 1189.

E-mail addresses: eva.manova@chem.ethz.ch (E. Manová), natalie.von.goetz@chem.ethz.ch (N. von Goetz), konrad.hungerbuehler@chem.ethz.ch (K. Hungerbuehler).

http://dx.doi.org/10.1016/j.envint.2014.09.008
0160-4120/

Available data on human exposure levels to EHMC via the use of PCPs are very limited (Andersen et al., 2012). Since EHMC is contained in multiple categories of PCPs that can be concurrently used by the same consumer, aggregate exposure estimates are highly needed for risk assessment. The major difficulty in modeling the aggregate consumer exposure to EHMC, particularly in European populations, is the lack of publicly available national-level data on use patterns for PCPs, including co-use data (i.e., which PCP categories are concurrently used by consumers), and exact EHMC concentrations in PCPs that are kept confidential in Europe.

EHMC is frequently contained in PCPs sold on the Swiss market (Manová et al., 2013b). Not surprisingly, therefore, it has been found in the Swiss environment in both abiotic (Fent et al., 2010; Plagellat et al., 2006; Poiger et al., 2004) and biotic matrices (Balmer et al., 2005; Fent et al., 2010), including breast milk (Schlumpf et al., 2010). The objective of the present study was to provide the very first comprehensive information about the current EHMC aggregate exposure levels through multiple PCP categories. Our novel probabilistic modeling method is performed at an individual level for the German-speaking Swiss consumers and is well suited for ingredients not present in all PCPs of a given product category, such as EHMC, to avoid unreasonable overestimation of exposure. Furthermore, in comparison to the conventional probabilistic modeling approaches, our approach takes into account correlations between frequencies of use of the different PCP categories used by the same consumer. Our work also explores the contribution of different PCP categories to aggregate exposure, as filling this major data gap is necessary to enable concerned consumers to minimize their exposure to EHMC via the use of PCPs.

2. Materials and methods

Despite its widespread use in PCPs, not all products in a given PCP category contain EHMC. To avoid overestimation of exposure, we assessed the aggregate exposure probabilistically at an individual level. For this purpose, two data sets were previously collected. As a first component of this work, we conducted a postal questionnaire survey to determine the use patterns of PCPs (Manová et al., 2013a). As a second component, we sampled 116 PCPs selected on the basis of the questionnaire and measured their EHMC concentrations (Manová et al., 2013b). For each respondent, we then linked the self-reported data on PCP use patterns with EHMC concentrations measured in the self-reported PCPs. Seven widely used leave-on PCP categories that are often used simultaneously were considered (face cream, aftershave lotion/balm, hand cream, makeup foundation, lip care, lipstick, sunscreen) for their high likelihood of containing UV filters.

2.1. Study design and data collection

First, we established a list of PCP categories that often contain UV filters. Based on our market analysis, eight target PCP categories have initially been chosen: face cream, body lotion, aftershave lotion/balm (hereafter referred to as aftershave), hand cream, makeup foundation, lip care, lipstick, and sunscreen. Other PCP categories (e.g., rinse-off products) may also contain EHMC as a UV absorber to prevent photodegradation of the product, but UV absorbers are usually only needed in very low concentrations. Usage data for the selected leave-on PCP categories were collected through postal questionnaires between January and March 2011. The study was conducted in the German-speaking part of Switzerland. Throughout this work, we use the term “children” for participants aged ≤12 years, “adolescents” for participants aged between 13 and 17 years, and “adults” for those ≥18 years old. Two questionnaire versions were designed; one for adults, and one for children and adolescents. The questionnaire for children and adolescents was mailed to 1000 eligible families recruited using a commercially available address database. The adult questionnaire was sent to 2500 household addresses randomly selected from the Swiss telephone directory. The overall response rate to the questionnaires was 48.8% for children and adolescents and 36.8% for adults. After exclusion of ineligible, incomplete, and contradictory responses, the final dataset included 1196 respondents (age range of ≤1–97 years). Detailed information about the questionnaire survey and the population characteristics have been published previously (Manová et al., 2013a). The present study was conducted as part of a larger research project focused on several organic UV filters, therefore ingredient lists of the frequently used PCPs identified by the questionnaire were inspected for the presence of all organic UV filters, not only EHMC. Body lotion was subsequently excluded from further consideration due to the absence of organic UV filters in the ingredient lists of body lotions used by the questionnaire respondents.

A sample of 116 UV filter-containing PCPs used by the respondents was selected for analysis. PCP samples were collected locally from several Swiss retailers (Migros, Coop, Denner, Lidl, ALDI SUISSE), local pharmacies and perfumeries in Zürich and Basel, Switzerland, and a Swiss internet retail store. Concentrations of 22 organic UV filters were measured in the selected PCPs. EHMC was detected in 59 products (51% of all PCPs tested). Details of the chemical methods and UV filter concentrations in the PCPs analyzed were described by Manová et al. (2013b).

2.2. Model background

The Ford model given in Eq. (1) (Ford, 1998; Wormuth et al., 2005) considers dermal application of liquid and viscous products and is thus well suited for characterizing EHMC exposure via the investigated PCP categories. For each individual in our study population, the Ford model was first used to derive the daily external exposure to EHMC, $E_{EHMC(external)}$ [mg day^{-1} kg^{-1}], from their self-reported PCP application frequency, f_{event} [day^{-1}], the retention factor of the considered PCP category (i.e., a measure of the fraction of EHMC-containing PCP that remains in contact with a human body over a longer period of time), a_{ret} [−], the amount of the EHMC-containing PCP applied per application, q_{PCP} [mg], and the weight fraction, w_{PCP} [−], of EHMC in the PCP applied by the respondent. Finally, the exposure values were adjusted for the respondent's self-reported body weight, m_{bw} [kg]. The daily internal exposure, $E_{EHMC(internal)}$ [mg day^{-1} kg^{-1}], was estimated by introducing the absorption factor, *abs* [−], representing the fraction of the external aggregate dose of unmetabolized EHMC that is transferred into the systemic circulation.

$$E_{EHMC\ (internal)} = f_{event} \cdot a_{ret} \cdot q_{PCP} \cdot w_{PCP} \cdot m_{bw}{}^{-1} \cdot abs \quad (1)$$

If a respondent was exposed to EHMC via more than one PCP category, then the aggregate exposure was defined as the sum of EHMC exposures across the different PCP categories. In accordance with our individual-oriented approach, each questionnaire respondent was taken as a starting point and the daily EHMC internal aggregate exposure from the different PCP categories was aggregated at an individual basis, while maintaining the information about each respondent's self-reported PCP co-use patterns, as given in Eq. (2), where n represents the number of PCP categories used by the respondent.

$$E_{EHMC,agg(internal)} = \sum_{i=1}^{n} E_{EHMC,PCP_i(internal)} \quad (2)$$

We computed the aggregate exposure separately for summer/autumn (May to October) and winter/spring (November to April), as the frequency of sunscreen use and the extent of application to different areas of the body both depend on the outdoor activities associated with the respective season. However, we have also derived the acute internal aggregate exposure to EHMC to account for the very intense exposure to EHMC related to sunscreen whole-body use on a sunbathing day