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Levels, variability and determinants of environmental phenols in pairs of Norwegian mothers and children

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ABSTRACT

Background: Exposure to environmental phenols including parabens, bisphenols (BPs), oxybenzone/benzophenone-3 (BP-3) and triclosan (TCS) is ubiquitous. Due to evidence of their estrogenic activity, they have been considered as chemicals of concern. The exposure of the Norwegian population to these compounds is presently unknown.

Aims: To measure urinary levels of twelve different environmental phenols including four emerging bisphenols: S, F, B and AF (abbreviated as BPS, BPF, BPB and BPAF, respectively) in a healthy Norwegian population. We have calculated short-term variability, estimated daily intakes and investigated important determinants of exposure.

Methods: Urine samples were collected from mothers (n = 48) and their children (n = 56) during spring/summer 2012 in two counties in Norway.

Results: Six environmental phenols namely methyl, ethyl and propyl paraben, BPA, BP-3 and TCS were detected in almost 100% of the urine samples. Among the emerging bisphenols, BPS was detected most frequently in the urine samples (42–48%) followed by BPF (4–15%). Parabens were positively and significantly correlated to each other in both mothers and children. Levels of parabens and BP-3 were higher in mothers compared to children. All mothers and children had lower estimated daily intakes (back calculated from the urinary concentrations) of parabens and BPA than the respective acceptable and tolerable daily intakes (ADIs and TDIs) established by the European Food Safety Authority (EFSA). Observed intraclass correlation coefficients (ICCs) indicated moderate to high reliability of spot urine measurements for all the environmental phenols (ICCs: 0.70–0.97). Use of hair products, deodorants, face and hand creams were significantly associated with higher urinary levels of parabens.

Conclusions: Occurrence of environmental phenols in healthy Norwegian women and children is abundant. Among emerging bisphenols, there is widespread exposure to BPS. A single spot urine sample can be used for estimating short-term exposures of environmental phenols. Urinary levels of parabens were associated with use of PCPs.

1. Introduction

Parabens, bisphenols (BPs), oxybenzone/benzophenone-3 (BP-3), triclosan (TCS) and triclocarban (TCC) are man-made chemicals used in various consumer products. Parabens are alkyl or aryl esters of *para*-hydroxy benzoic acid and have mainly been used as antimicrobial preservatives in food, personal care products (PCPs) and pharmaceuticals (Boberg et al., 2010). Bisphenol A (BPA) is used in manufacturing of polycarbonate plastic and epoxy resins and is found in different products like cans (food and drink), dental sealants, thermal receipts, food packaging and PCPs (Geens et al., 2012). In addition, emerging bisphenols like BPS, BPF, BPAF, BPAP, BPB, BPZ, BPP and bisphenol A

Abbreviations: ADI, acceptable daily intake; BMI, body mass index; BW, body weight; BP, bisphenol; BP-3, oxybenzone/benzophenone-3; BuP, butyl paraben; DEMOCOPHES, Demonstration of a study to Coordinate and Perform Human biomonitoring on a European Scale; EDCs, endocrine disrupting chemicals; EFSA, European Food and Safety Authority; EtP, ethyl paraben; F_{ue}, fraction excreted in urine; ICC, intraclass correlation coefficient; GM, geometric mean; LOD, limit of detection; LOQ, limit of quantification; MeP, methyl paraben; MoBa, Mother and Child Cohort study; MRM, multiple reaction monitoring; MP, mobile phase; MS, mass spectrometry; NA, not analyzed; ND, not detectable; NOAEL, no observed adverse effect level; PCPs, personal care products; PrP, propyl paraben; RSD, relative standard deviation; SCCP, Scientific Committee on Consumer products; SG, specific gravity; SPE, solid phase extraction; ln, natural logarithm; TDI, tolerable daily intake; TCS, triclosan; TCC, triclocarban; UPLC, ultra-performance liquid chromatography; UV, ultraviolet

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diglycidyl ether are used as substitutions for BPA in some of the consumer products (Gramec Skledar and Peterlin Masic, 2016). BP-3 is mainly used as UV filters in sunscreens and as UV stabilizer in some food packaging (Krause et al., 2012). TCS and TCC are used as antimicrobial and antifungal agent in products like toothpaste, soaps, detergents and other hygiene and PCPs (Witorsch and Thomas, 2010). Apart from TCC, all these compounds have a phenol group in their chemical structure and are often referred to as "environmental phenols" as they are known to be widespread in the environment.

Although these environmental phenols are non-persistent chemicals and have short elimination half-lives in humans (parabens: 1–7 h, bisphenols: 6 h, BP-3 and TCS: < 24 h) (Kadry et al., 1995; Kim and Choi, 2014; Moos et al., 2016a; Sandborgh-Englund et al., 2006; Thayer et al., 2015), their widespread use and potential endocrine disrupting properties have made them chemicals of concern (Bergman et al., 2013; Ghazipura et al., 2017). Due to its estrogen activity and reproductive toxicity, BPA has been banned in manufacture of infant feeding bottles in Europe since 2011 (EFSA Panel on Food Contact Materials, 2015). In a recent risk assessment of BPA published by European Food and Safety Authority (EFSA), the tolerable daily intake (TDI) was decreased from 50 μg/kg body weight (bw) to 4 μg/kg bw (EFSA Panel on Food Contact Materials, 2015). Exposure to parabens, BP-3, TCS and TCC have been associated to weak estrogenic activity and some adverse health effects in humans (Bledzka et al., 2014; Giulivo et al., 2016; Kim and Choi, 2014; Wang and Tian, 2015). EFSA has established an acceptable daily intake (ADI) of < 10 mg/kg bw for the sum of methyl paraben (MeP) and ethyl paraben (EtP) (EFSA, 2004). Scientific committee on consumer products (SCCP) has regulated the use of BP-3 and the maximum allowed concentration is 6% in ready for use preparations (SCCP, 2008). The use of TCS is also regulated and is allowed in some cosmetic products up to a concentration of 0.3% (SCCP, 2009).

In Norway, only two studies presenting levels of MeP, propyl paraben (PrP), butyl paraben (BuP), BPA and TCS in 45 Norwegian pregnant women are available (Bertelsen et al., 2014; Guidry et al., 2015). There is only one study available showing TCS levels in Norwegian children (Bertelsen et al., 2013).

Thus, the aim of the present study was to determine the levels of 12 different environmental phenols (four parabens (MeP, EtP, PrP and BuP), five bisphenols (BPA, BPS, BPF, BPB and BPAF), BP-3, TCS and TCC) in Norwegian healthy mother-child pairs. Further, we have studied the diurnal variation and calculated the intraclass correlation coefficients (ICCs) in mothers. The determinants of exposure of these urinary phenols (diet and PCPs) were also investigated in the present study.

2. Materials and methods

2.1. Study subjects and sample collection

The study comprised 48 mothers and 56 children as described elsewhere (Sakhi et al., 2017). In brief, 48 mother-child pairs from 2 different Norwegian counties participated in the study. In addition, siblings of 8 children were also included in the study (in total 56 children). The sampling period was one complete day (24 h) and for most of the participants, it started during the evening of the first day and ended the consequent evening next day (Sakhi et al., 2017). The mothers were encouraged to collect all the spot urine samples during the study period of 24 h. Each spot urine sample was collected in a new container and labelled with day and time of collection. All the mothers provided first morning urine sample. Among 48 mothers, 21 (44%) provided all the spot urine samples, 13 (27%) collected > 3 spot urine samples and 14 collected 2–3 spot urine samples, resulting in a total of 244 spot urine samples. Among 56 children, 54 collected first morning and 53 collected afternoon/evening spot urine samples (in total 114 spot urine samples). The urine sample containers used in the present study were made of high-density polyethylene (HDPE) and the blank samples prepared in these containers showed no detectable concentration of phenols. The samples were stored in a freezer at −20 °C until analysis.

2.2. Analytical method and QA/QC measures

The environmental phenols were determined using on-line solid phase extraction (SPE) prior to ultra-high performance liquid chromatography (UPLC) coupled to tandem mass spectrometry (MS-MS). The present method included 4 parabens (MeP, EtP, PrP and BuP), 5 bisphenols (BPA, BPS, BPF, BPB and BPAF), BP-3, TCS and TCC. This analytical methodology is a modification of the previously published method (Zhou et al., 2014) and is fully validated for the above-mentioned phenols. In brief, labelled internal standards and enzyme solution were added to 200 μL of the sample. After 4 h, the enzymatic reaction was stopped by adding formic acid, the samples were centrifuged and 80 μL of the supernatant was injected into the UPLC-MS-MS system. The ionization of the analytes was performed in an electrospray source in negative mode. A signal to nose (S/N) ratio of 10 was considered as limit of quantification (LOQ). The LOD (S/N ratio of 3) was calculated from the respective LOQs and varied from 0.02–0.10 ng/mL (Table S2). Further details of the method are described in the supplementary information and Tables S1–S2.

The validation was performed at 5 different concentration levels (from 0.2 to 600 ng/mL) obtaining inter and intra precisions lower than 34% and accuracies between 69 and 154% (Table S3 and S4). The validation results were satisfactory for the environmental phenols as demonstrated by the low relative standard deviation (RSD < 26%) obtained using in-house controls and National Institute of Standards and Technology (NIST) reference material (Table S5). Additionally for BPA and TCS, two different inter-laboratory comparisons showed low z-score (between −1.30 and 0.09) and concentrations within the tolerance range (Table S6).

In order to correct for the urinary dilution, both creatinine and specific gravity (SG) were measured in all the spot urine samples as described previously (Sakhi et al., 2017). The SG adjusted concentrations were used in all the statistical analysis, because SG concentrations are less effected by age, gender, BMI, muscle mass, diet, activity and season compared to creatinine concentrations (Johns et al., 2015). The unadjusted and creatinine adjusted concentrations are presented in the supplementary information (Tables S7 and S8).

2.3. Statistical analysis

IBM SPSS version 23 and Stata (StataCorp LLC, Texas, USA) were used for the statistical analyses. The urine samples were grouped into 3 time-periods based on likely indoor exposure and daily routine activities (evening: 16–24 h, morning: 24–8 h and day: 8–16 h) (Sakhi et al., 2017). Many of the environmental phenols are used in different PCPs, which are used mostly in the morning. Thus, the day time-period samples was further divided into two time-periods of 4 h each (early day: 8–12 h and afternoon: 12–16 h) (Sakhi et al., 2017). For children, the urine samples were grouped into two time-periods (morning: 24–8 h and afternoon/evening: 12–18 h) (Sakhi et al., 2017). The mean of the concentration was taken if the participant had more than one urine sample in the specified time-period (number of participants with more than one urine sample in time periods: first evening 28, morning 9, early day 4, afternoon 15 and second evening 1). Wilcoxon test was used to compare (i) environmental phenol concentration between morning time-period (reference) and other time-periods in mothers (ii) environmental phenol concentrations in the morning urine samples with afternoon samples among children and (iii) environmental phenol concentrations in mothers' morning urine samples with the corresponding children samples. Spearman rank test was used to study correlations between different environmental phenols in both mothers and children. The statistical analysis in (ii) and (iii) were done with the

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