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Research article

Sampling strategy for estimating human exposure pathways to consumer chemicals

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ABSTRACT

Human exposure to consumer chemicals has become a worldwide concern. In this work, a comprehensive sampling strategy is presented, to our knowledge being the first to study all relevant exposure pathways in a single cohort using multiple methods for assessment of exposure from each exposure pathway. The selected groups of chemicals to be studied are consumer chemicals whose production and use are currently in a state of transition and are; per- and polyfluorinated alkyl substances (PFASs), traditional and “emerging” brominated flame retardants (BFRs and EBFRs), organophosphate esters (OPEs) and phthalate esters (PEs). Information about human exposure to these contaminants is needed due to existing data gaps on human exposure intakes from multiple exposure pathways and relationships between internal and external exposure. Indoor environment, food and biological samples were collected from 61 participants and their households in the Oslo area (Norway) on two consecutive days, during winter 2013-14. Air, dust, hand wipes, and duplicate diet (food and drink) samples were collected as indicators of external exposure, and blood, urine, blood spots, hair, nails and saliva as indicators of internal exposure. A food diary, food frequency questionnaire (FFQ) and indoor environment questionnaire were also implemented. Approximately 2000 samples were collected in total and participant views on their experiences of this campaign were collected via questionnaire. While 91% of our participants were positive about future participation in a similar project, some tasks were viewed as problematic. Completing the food diary and collection of duplicate food/drink portions were the tasks most frequent reported as “hard”/”very hard”. Nevertheless, a strong positive correlation between the reported total mass of food/drinks in the food record and the total weight of the food/drinks in the collection bottles was observed, being an indication of accurate performance of the participants despite the challenges of the sampling campaign.

1. Introduction

In our everyday life we use many consumer products that contain a range of chemicals added to meet regulatory requirements, such as fire retardancy. Direct and/or indirect contact with such products can result in human exposure to these “consumer chemicals”. Information on human exposure pathways is essential for identification of high risk population sub groups and for the development of efficient control strategies to minimize

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human exposure. It is therefore of high importance to identify the major human exposure pathways and quantify intakes for in-use substances, phased-out substances and their replacements. Among such chemicals are per- and polyfluorinated alkyl substances (PFASs), brominated flame retardants (BFRs), organophosphate esters (OPEs) and phthalate esters (PEs). These selected groups of organic chemicals are of interest owing to concerns about their potential for human exposure and consequent adverse health effects [1]. Some substances in these groups have been phased out of production and replaced with either 1) substances in the same group or 2) structurally unrelated substances outside of the group. As a result of the changes in chemical production and use patterns, the chemicals we are exposed to, the pathways of exposure as well as the intakes of chemicals are changing and need to be monitored. Further, these chemicals provide exemplars for other groups of contaminants with similar physical–chemical properties and applications.

PFASs are anthropogenic chemicals used since the 1950s in many industrial and consumer products, such as surfactants, water and oil repellents, varnishes, waxes, lubricants, hydraulic oils and fire-fighting foams [2]. BFRs are present in a broad range of commercial products, including electronic devices, textiles, carpeting, building insulation and furniture [3]. PEs are high-production-volume chemicals used mainly as plasticizers and can be found in a wide range of consumer products such as plastic toys, personal care products, paints and pharmaceuticals [4]. Recent restrictions in the production and use of some of the chemicals aforementioned, like polybrominated diphenyl ethers (PBDEs) and perfluorooctanesulfonate (PFOS) have led to the production and use of alternatives. Among such chemicals are the so-called emerging brominated flame retardants (EBFRs) and OPEs. OPEs are either non-halogenated or halogenated. The non-halogenated congeners are mostly used as plasticizers in several consumer products, while the halogenated congeners are mostly used as flame retardants in a range of products including textiles, rubber, polyurethane foam, antistatic agents, cotton and electronic equipment [5].

Human exposure to the selected groups of chemicals can occur through food consumption and drinking water. Exposure can also ensue following their release to the indoor environment, via air inhalation, dust inhalation and ingestion, direct hand contact with consumer products followed by hand-to-mouth contact, and dermal absorption [6–10]. The relative magnitude of these pathways varies, being dependent on several source-related, physical–chemical, environmental and human behavioural factors. Regional differences can also be observed for the relative importance of exposure pathways for the same chemical class. For example, in some studies dietary intake has been suggested as the main exposure pathway for some PBDEs, while in other studies, indoor dust has been identified as the major source of exposure for PBDEs [11–14]. However, for many of the above-mentioned classes of chemicals, the relative significance of different human exposure pathways is not well characterized, while exposure studies have in the past tended to focus on one or a few exposure pathways at a time.

The sampling strategy and design is a key step in order to achieve desirable and reliable results when assessing human exposure. There are two main ways of performing an exposure assessment; 1) measure the external exposure (i.e. the total intake from multiple exposure pathways) or 2) measure the internal exposure (i.e. the body burden). Further these assessments can be conducted using different sampling designs. A cross sectional sampling study involves sampling multiple representative participants and their exposure pathways at one time point to provide a snapshot of external and/or internal exposure [6]. A cross-sectional study can be repeated several times (usually using different participants) to determine how exposure of different population groups has changed over time [15–17]. A longitudinal sampling study involves sampling the same participants/exposure pathways at several time points (e.g. repeatedly sampling the same individuals during their lifetimes) to determine how the exposure of these individuals changes with time [18–23]. These different designs have strengths and limitations as described in Table 1. In addition, human exposure can be indirectly estimated through mathematical modelling, but this is not the focus of our study [24].

In the present study we conducted a comprehensive cross-sectional study collecting samples representative for both external and internal exposure. This design was considered the most appropriate for our purposes as it is time and cost effective and at the same time allows (a) evaluation of the relative importance of different external human exposure pathways, (b) comparison of different external exposure metrics regarding their ability to reflect participants' body burdens, and (c) assessment of the suitability of invasive and non-invasive samples for biomonitoring purposes. The sampling campaign was carried out in Oslo, Norway with the objective of collecting a wide variety of biological, food and environmental samples from a cohort of participants and their homes, using several sampling approaches undertaken simultaneously. To our knowledge this is the first study that has collected samples from all relevant exposure pathways using multiple methods for assessment of exposure of each pathway. The aim of this paper is to provide a detailed description of the methods employed for the collection and processing of samples of the cohort, that can be used in future studies. Also discussed are practical and scientific aspects, ethical issues, as well as the limitations and uncertainties of the sampling campaign and how they can be minimized. Moreover, we also report participant's views on their experiences of their participation in the campaign to inform design of similar future campaigns so that participant recruitment and completion rates are maximized.

2. Materials and methods

2.1. Study population and overview of sampling

This sampling campaign was conducted as part of the A-TEAM project. The project's objective is to enhance knowledge and substantially improve the approaches currently used to identify and monitor external and internal human exposure to consumer chemicals; specifically PFASs, EBFRs, OPEs and PEs. It also aims to provide robust information on the relative importance of different exposure pathways to our target contaminants and reduce the gaps between external and internal dose.

To achieve our objective, we established a study group of 61 Norwegian adult males and females from Oslo, Norway. Study participants were recruited from the staff of the Norwegian Institute of Public Health (NIPH). During the recruitment, all NIPH employees were invited to participate by an electronic announcement published at the institute's website accessed by NIPH employees only. While we recognize that participants recruited might not be a representative sample of the overall Norwegian population, we consider our study population a fit-for-purpose "convenience sample" that is both easy to reach and communicate with, and for which logistical issues are minimized. In addition, a comprehensive sampling campaign like this requires motivated participants in order to get reliable results. Sample collection was conducted during the winter period when the proportion of time spent indoors is at a maximum and ventilation is at its minimum. To characterize as many exposure pathways as possible, samples relevant to both external and internal exposure were collected (Fig. 1). Sampling