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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

Table 1
Characteristics, strengths and limitations of different sampling designs for assessing human exposure.

		Characteristics	Strengths	Limitations
External dose		Measure concentrations of relevant chemicals in different exposure media and combine with exposure factors (e.g. inhalation rates)		
	Cross-sectional [6]	Observational study at multiple locations carried out at one time point ("snapshot") or over a short period. This can be repeated at multiple time periods, but not at the same locations.	- Provides information on the relative importance of different external exposure pathways at a given time point. - Project does not have to be long-term. - Because a cross-section of the population can be sampled, differences in age, gender, ethnicity etc. can be taken into account.	- If undertaken only once, provides no information on changes in exposure over time - Cannot relate external and internal exposure for chemicals with long half-lives
	Longitudinal [18]	External exposure studies repeated at different time points, but at the same multiple sampling stations in all studies. The aim would be to follow the external exposure for the same individuals (e.g. in homes or occupational exposure settings).	- Provides information on the relative importance of different exposure pathways at several time points for the same sampling stations - Provides information on changes in exposure over time for given individuals - Desirable design when comparing external dose with internal dose for chemicals with long half-lives	- Expensive - Time consuming; the project must be long-term (typically >>5 years) - Does not take individual differences into consideration (e.g. age and gender) - Challenging to have high number of participants
Internal dose (biomonitoring)		Measure concentrations of relevant chemicals in a biological matrix and combine with knowledge on distribution and elimination. Common to use questionnaires to evaluate associations between measured concentrations and factors affecting exposure (e.g. consumption of fish)		
	Cross-sectional [6,15]	Observational study for multiple individuals carried out at one time point ("snapshot") or over a short period. This can be repeated at multiple time periods, but not for the same individuals (a representative population is normally sampled).	- Reflects an integrated exposure over time (depending on the half-life) comprising various sources and pathways - Can take individual differences into consideration (e.g. age and gender) - Often high number of participants - Frequently used for assessing relationships between exposure and health outcomes	- Provides limited information on the relative importance of different exposure pathways unless (1) it is combined with an external dose cross-sectional study (e.g. this present study) or (2) there are good bio-markers of external exposure pathways - Provides no information on exposure changes over time unless repeated at regular intervals (e.g. NHANES). Even then can only show temporal exposure changes on the population level and not on the individual level.
	Longitudinal [19–22]	The same participants are sampled in all studies and followed over time (e.g. over lifetime).	- Shows how exposure changes over time for individuals from various sources and pathways - Takes individual differences into consideration (e.g. age, gender and specific exposure behaviour) - Desirable design when comparing external dose with internal dose for chemicals with long half-lives - Highly suitable for assessing relationships between exposure and health outcomes	- Provides limited information on the relative importance of different exposure pathways unless (1) it is combined with an external dose longitudinal study or (2) there are good biomarkers of external exposure pathways - Expensive - Time consuming; the project must be long-term (typically >>5 years) - Demanding for participants who are monitored over many years and at different phases of their life - Often limited number of participants and many drop out or are excluded during a long-term study

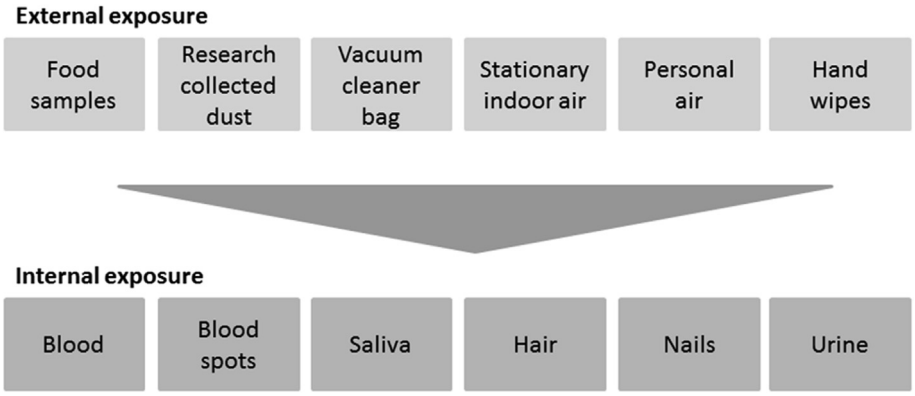


Fig. 1. Rationale of human exposure and sample collection.

of each participant occurred over 2 days during 2 visits by 2 researchers to the participant's house (1 visit per day). A schematic overview of events conducted over the 2-day sampling period is provided in Fig. 2. Some samples, such as urine, food samples, saliva, nails and hand wipes were collected by the participants themselves, in accordance with detailed written instructions provided by the sampling team. Other samples, such as house dust and blood spots were collected by sampling researchers. Detailed information about dietary habits, the indoor domestic environment and other lifestyle characteristics of the participant was collected through questionnaires.

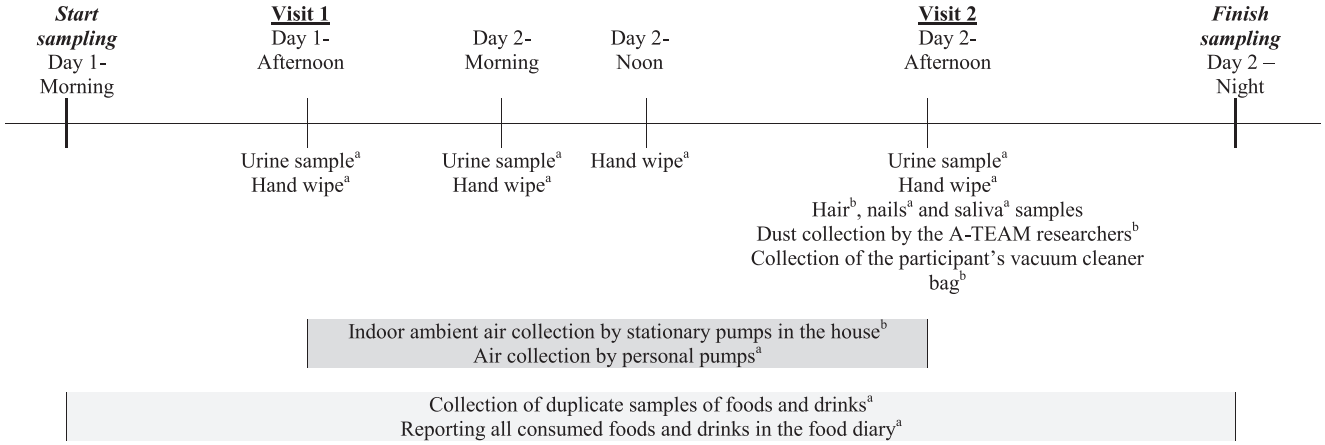
The A-TEAM sampling campaign was approved by the Regional Committees for Medical and Health Research Ethics in Norway (2013/1269), and all participants completed a written consent form before participating.

2.2. Collection of home air samples

Indoor air samples were collected by connecting sampling media specific for different classes of target contaminants to three SKC Leland Legacy low volume pumps (SKC Inc., Eight Four, PA, USA) programmed to sample for 1440 min (24 h) and placed in a fixed point in participants' living rooms. The pumps were installed as far away as possible from doors, windows, stoves and display screens that were in use during the 24 h sampling. Sampling media used were: for PEs and OPEs, four parallel ENV + cartridges (0.2 g, 6 mL) (Biotage, Charlotte, NC, USA); for PFASs, four parallel ENV + cartridges (0.2 g, 6 mL) [25–27], and for EBFRs, four parallel sampling trains containing two polyurethane foam plugs (PUFs) and one glass fibre filter (GFF) [28]. The flow rates were 5 L min^{-1} for PEs, OPEs and PFASs, and 12 L min^{-1} for EBFRs. We collected four non-used cartridges or sampling trains as field blanks, by connecting the train/cartridge to the pump tubing and exposing it to the indoor environment of each house for 5–10 s, with the pump turned off. After sampling, all cartridges and the field blanks were collected, wrapped in aluminium foil, and stored at −22 °C in labelled ziplock plastic bags.

2.3. Collection of personal air samples

Personal air samples were collected using one low volume SKC pump 224-PCMTX4 (SKC Inc., Eight Four, PA, USA) at a flow rate of 1 L min^{-1} for all contaminant classes. The pump was housed in a



*Blood samples and blood spots were collected before or after the 2-days sampling period, depending on the convenience of the participants.

[a] Samples collected by the participants

[b] Samples collected by the researchers

Fig. 2. Schematic overview of the 2-days sampling in the A-TEAM project.

backpack that accompanied participants throughout the 24 h sampling period. The sampler was attached to the participant's shoulder and participants were advised to keep the sampler close to their face during the entire 24 h sampling event, including sleeping hours. Since one pump was carried to collect personal air samples, only one sampler was connected. In contrast to the sampling of indoor air via stationary samplers, for which all contaminants groups were monitored for all participants, for personal air each participant collected one sample that was analysed for one contaminant group only. Hence, for PEs, OPEs and PFASs, an ENV + cartridge (1 g, 25 mL) was used, while for EBFRs, one sampling train containing two PUFs and one GFF was used. The cartridges for collecting air samples for OPEs and PEs analysis were pre-cleaned with acetone [27]. One cartridge or sampling train was collected for each participant, as a field blank for personal air following the same procedure as for stationary air field blanks. The same storage procedure was also followed for the collected samples and field blanks. Collected samples and field blanks were stored at 22 °C.

2.4. Collection of indoor dust samples

All participants were contacted 2–3 weeks before sampling and advised not to vacuum the living room until after the researchers' visit, who will perform the dust sampling. Dust samples from the whole floor and from all available elevated surfaces >0.5 m above the floor in the living room (i.e. windows, doors, picture frames, shelves, upholstered and non-upholstered furniture, books) were collected separately from each household using a vacuum cleaner equipped with a forensic nozzle and a one-way filter housing [29]. The time of vacuuming and the area of the living room that was vacuumed were also recorded. Clean forensic nozzle and filter were used between samples. After sampling, the filters were wrapped in aluminium foil and stored at 4 °C. One field blank was collected in each house by connecting an unused filter in the vacuum cleaner and exposing it in the indoor environment for some seconds.

In order to sub-divide collected dust samples for analysis, collected samples were aliquoted. Before aliquoting, any kind of visible undesirable substances in samples, such as hair and small food pieces that might contaminate our samples, were removed using pre-cleaned stainless steel tweezers and a spatula. No further sieving was conducted. Afterwards, settled and floor dust samples were divided directly from the filter (in order to avoid sample looses) into four subsamples each, by weighing on an analytical balance. Additionally, the filters from the field blanks, the floor dust sample and the settled dust sample were each cut in four parts and placed in separate containers. The subsamples prepared for PEs, OPEs and PFASs analysis were stored in polypropylene (PP) containers, while all subsamples prepared for EBFR analysis were stored in pre-cleaned, amber glass containers to minimize photodegradation.

Participants were advised not to discard their vacuum cleaner bag and to provide it to the researchers at the end of the 2-day sampling event. Collected bags were wrapped in Al foil, placed in a plastic bucket and stored at room temperature. Subsequently, a 500 μm sieve was used to sieve the dust from the bag. Six 2 g aliquots of the sieved dust were prepared in 30 mL containers and one 2 g aliquot in a glass tube wrapped in Al foil.

2.5. Collection of hand wipes samples

Four pairs of hand wipes were collected by each participant during the 2-day sampling period; one during the first visit, one on the morning of the next day, one at noon and one during the second visit. In order to collect all samples in an identical fashion, researchers demonstrated the hand wipe collection at the first home visit. Participants were advised not to wash their hands 60 min before collecting every hand wipe. The first hand wipe sample was reserved for PE analysis and was collected by immersing a piece of glass wool in 3 mL of isopropanol and wiping both sides (palm and back) of one hand from wrist to fingertips. Another piece of glass wool was used to wipe the other hand [30]. Both pieces of glass wool were stored in a 125 mL PP container. The glass wool was combusted in an oven at 450 °C for 24 h prior to sampling in order to remove possible contamination. The same sampling procedure was used for OPEs, PFASs and EBFRs except that gauze pads (Sterile Gauze Pads, 3 × 3 inches, Swift First Aid Inc. Valencia, CA, USA) were used instead of glass wool. For EBFRs, the gauze pads were stored in 60 mL amber glass containers after sampling.

2.6. Collection of duplicate diet samples and the food diary

In the duplicate diet method, a duplicate portion of all foods and drinks consumed over 24 h was collected. After receiving detailed guidance, participants were asked to collect weighed duplicate food and drink portions prepared as for consumption over 2 consecutive days. In addition, they received a kitchen scale capable of weighing food samples to ±0.1 g, four 2 L PP bottles, and a food diary. Participants were instructed to collect solid and liquid food samples in different 2 L bottles for each day. After collection, solid food samples were weighed, transferred into a food processor (Robot-coupe Blixer 3) and blended for 2–5 min. After homogenization of each sample, six subsamples of 100 g were prepared in 250 mL PP bottles and one in a 250 mL pre-cleaned amber glass jar. Liquid samples were homogenized by hand shaking and the same aliquoting procedure was followed. The ability of the food processor to homogenize the collected solid food samples was tested beforehand.

During collection of food and drink, the participants had to weigh and record all items that were prepared as duplicates in the food diary. As is common in food diaries, information on type, amount and time of consumption was reported for each consumed item. The unique component of our food diary is the addition of questions about packaging material (plastic box/bag/wrap, aluminium foil, paper/carton, and original package), cooking/preparation method (cooked, fried, raw washed or unwashed), the cooking utensils used (utensils with non-stick coating or other, microwaved in plastic or other) and serving vessel (paper/plastic/glass/porcelain cup or plate).

2.6.1. Testing the ability of the food processor to homogenize composite solid food samples

A composite food sample was collected and 10.7 g of NaCl was added into a portion of 216.7 g of the food sample. This was homogenized using the food processor, and then the entire food sample was added to the food processor and processed for 5 min. Five subsamples of 10 g from the processed food sample were transferred to glass containers. 100 mL of ionized water was added to each and were placed in ultrasonic bath for 10 min. Afterwards, the samples were centrifuged (2500 rpm for 15 min) and filtered. The conductivity was measured 3 times in each sample by a conductivity meter (Bench pH/mV/°C meter pH 1000 L, pHenomenal®). The relative standard deviation of the conductivity was 3.3, 3.1 and 2.5%, respectively, and the sample was considered sufficiently homogeneous since the relative standard deviation of the conductivity was below 3%. Hence, the food processor (Robot-coupe Blixer 3) sufficiently homogenized composite food samples.

2.7. Questionnaires

A food frequency questionnaire (FFQ) was also employed. This FFQ was designed and used for dietary assessment in the Norwegian Mother and Child Cohort Study (MoBa), and its modified version has been used in other studies of Norwegian adults [31,32]. This is a validated, semi-quantitative, self-administered FFQ where participants are asked to report their average frequency of intake of 255 food items for the last year. The frequency intervals ranged from never, to more than eight times per day, while portion sizes were given as units of fruits, slices of bread and cups/glasses of drinks.

An indoor environment questionnaire was designed to collect lifestyle and socio-demographic characteristics and habits of the participants, which could influence external and/or internal exposure to our target contaminants. These included: weight, height, number of deliveries and breastfeeding history for women, education, age, use of a car, use of house cleaning products, use of materials at work, use of personal care products etc. Additional information about the household was also collected, such as number of people living in the house, number of rooms, location of the house, description of materials used in the house and the furniture, numbers of electronic equipment, recent renovations, etc.

2.8. Collection of biological samples

A venous blood sample was collected from each participant by a research nurse at the NIPH during a scheduled appointment. Around 50 mL of whole blood was collected in four 10 mL plastic BD Vacutainer® serum tubes, to provide serum, and in one 10 mL plastic BD Vacutainer® whole blood tube with EDTA, to provide whole blood and plasma. Whole blood was left to clot for 1 h. Later, serum was separated by centrifugation at 2200–2500 rpm for 15 min. The collected serum was transferred to a 30 mL PP bottle and four subsamples of 2 mL were prepared in 2 mL PP cryogenic vials as well as one subsample of 4 mL in a 15 mL glass tube. Additionally, 2 mL of the collected whole blood were transferred to 2 mL PP cryogenic vials for storage and the rest was centrifuged at 2200–2500 rpm and plasma was transferred to a 10 mL tube. Blood spots were collected by pricking the inside tip of a finger of the non-dominant hand of the participant with a hand punch (BD Microtainer® Contact-Activated Blue Lancet) and applying blood drops on 2 blood spot cards (Whatman® protein saver cards). The used cards and unused blank cards, to be used as field blanks, were left to fully air dry horizontally overnight at room temperature and put into a foil-barrier bag with 2 desiccant packs.

Participants were asked to collect 3 urine samples over the 2 day sampling campaign; one in the afternoon of the 1st day, one in the morning of the 2nd day and one in the afternoon of the 2nd day. Three 500 mL high-density polyethylene (HDPE) bottles with screw caps and security lids were provided in advance. The participants noted on the bottle the date and time of each urine collection and kept the samples in a cool place until passed to the researcher team. Subsequently, urine samples were weighed and divided into 10 mL subsamples.

Collection of hair samples was based on the COPHES protocol [33]. In brief, for participants with long hair length (>3.5 cm), several strands of hair were collected horizontally and rolled to form a lock. A piece of adhesive tape was placed in the upper part of the hair lock and was used to fasten the hair lock. The lock was cut as close to the scalp as possible and placed in a paper envelope. Two hair locks were collected, one from each side of the head. For participants with short hair (<3.5 cm), several strands of hair were cut from the sides of the head and were collected in a paper envelope. The hair sample under the adhesive tape will not be used for chemical analysis. Additionally, participants were asked not to cut their fingernails 2–3 weeks prior to the first scheduled home visit. Fingernails were collected as one composite sample per participant in a paper envelope between the two home visits. Participants were also advised to remove any nail polish, dirt, debris and artificial nails before clipping their fingernails. Saliva samples were collected as 5 spits in a 60 mL PP wide-mouth bottle during the second scheduled home visit.

All samples were stored at −20 °C until delivery to the project partners.

2.9. Evaluation of participants' experiences of the sampling campaign

In order to record the experience of the participants during the sampling campaign, a short form, rating their views on various aspects of the sampling campaign, was developed and administered. The form, sent to the participants after they had completed the sampling, was completed anonymously and returned to the project leader. This rating form included 5 questions: gender, future participation in case of a similar project (yes/no), the motivation to participate in the A-TEAM study, rating of all the tasks performed during their participation and evaluation of the tasks performed during the participation. The last question was included in order to assess possible reasons for any difficulties that participants had during their participation. The form is included in the supplemental material.

2.10. Statistical analysis

Descriptive statistics were used for continuous lifestyle and socio-demographic characteristics of the participants and the masses of collected samples, frequencies were used to summarize categorical variables. Differences in collected masses were tested by t-test for paired samples of urine and the non-parametric Wilcoxon signed rank test for paired samples of foods, drinks and dust. Non-parametric tests and the non-parametric Spearman's rank correlation coefficient (rho) were used in case of continuous variables with non-normal distributions as assessed by the Shapiro–Wilk test of normality.

3. Results

3.1. Home visits and participant characteristics

The sampling campaign was conducted between November 2013 and April 2014 and involved 122 home visits to 61 households. Visits were scheduled according to the participant's convenience with a rate of 8 visits per week. Regarding household locations, 80% were located less than 20 km from the NIPH, with the rest located between 21 and 162 km distance. We obtained complete sample sets for 52 participants (85%). The most common reason for not having a complete set of samples was the lack of hair sample (n = 4 participants), while one participant provided 3 hand wipes instead of 4, one participant did not provide nails, 2 participants did not have a vacuum cleaner and one participant did not provide nails and vacuum cleaner bag samples.

Forty-five participants (74%) were women and 28 of them had given birth (Table 2). The average age was 42 years (SD 11.3), the average weight was 71 kg (SD 15) and the average body mass index (BMI) was 24.2 kg/m^2 (SD 4.4). Most of our participants were highly educated (more than 12 years), were born in Norway and were non-smokers. All participants worked outside their houses.

Table 2
Characteristics of participants in the A-TEAM sampling campaign.

Characteristics	Average	(Standard deviation)	Minimum	Maximum
Age (years)	42	(11)	20	66
Weight (kg)	71	(15)	52	125
BMI (kg/m^2)	24.2	(4.4)	19.4	47.0
	N	%		
Gender				
Men	16	(26)		
Women	45	(74)		
Women with children				
No	17	(38)		
1 child	6	(13)		
2–3 children	22	(49)		
Education				
Low (≤9 years)	0	(0)		
Middle (10–12 years)	4	(7)		
High (≥12 years)	57	(93)		
Born in Norway				
Yes	44	(72)		
No	17	(28)		
Smoking status				
Never	43	(71)		
Currently smoking	1	(2)		
Sometimes/quit smoking	17	(27)		

3.2. Biological samples

All participants provided blood and urine samples. From the 40 mL of venous blood, 12 mL (±1.5 mL) of serum was obtained per participant on average (30% of the venous blood volume, min–max = 15–38%, Table 3). Blood collection was performed on average 4 days after the first scheduled visit at the participants' house. The morning urine (sample 2) was of significantly greater volume than the afternoon urine sample taken the same day (sample 3). The "non-invasive samples", blood spots, hair, nails and saliva, were collected from more than 97% of our study group (57 hair, 59 nail and 61 saliva samples, 116 blood spot cards).

3.3. Duplicate diet food samples

The daily weight of the solid food collected from each participant ranged between 0.3 and 1.8 kg, while the daily weight of liquid food was up to 4 kg (Table 3). The mass of food and drink collected on the first sampling day significantly exceeded that on the second day because on day 2 some of our participants collected samples up until the afternoon visit of the research team rather than for the whole day. Additionally, we found a positive and significant correlation between the mass of food collected over the 2 days (solid food: rho = 0.46, p-value <0.001 and liquid food: rho = 0.61, p-value <0.001), suggesting low individual variation of the total amount of consumed food within the two days. We also observed 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, and the difference between the collected and reported amount was not significant (Fig. 3). The separate positive correlations for solid and liquid foods were all significant and similar to the overall correlation, ranging from 0.77 to 0.94. Given that the collection of samples in a duplicate diet study and the completion of a food diary are both demanding and complicated tasks, the good agreement between the reported and collected food/drinks mass is an indicator of success of this duplicate diet sampling. We acknowledge that the high educational level and the involvement/occupation in the research field might have contributed to this good performance in the duplicate diet study.

Table 3
Masses of urine, serum, food and dust collected from participants during the A-TEAM sampling campaign.

Matrix	Average	(Standard deviation)	Minimum	Maximum
Biological samples				
Gross weight of urine (g)				
Sample 1	180	(87)	39	410
Sample 2	192	(91)	15	413
Sample 3	166	(81)	26	388
Serum volume (mL)	11.7	(1.5)	6.0	15.0
Food samples				
Weight of solid food (g)				
Sample 1	970	(361)	270	1841
Sample 2	689	(279)	159	1277
Gross weight of liquid food (g)				
Sample 1	1915	(736)	541	4030
Sample 2	1619	(754)	470	4254
Dust samples				
Weight of dust collected by the researchers (g)				
Elevated surface dust	0.59	(0.38)	0.12	2.02
Floor dust	1.07	(1.01)	0.11	6.39
Dust from participant's vacuum cleaner bag (g)	67.1	(82.6)	1.0	380.1

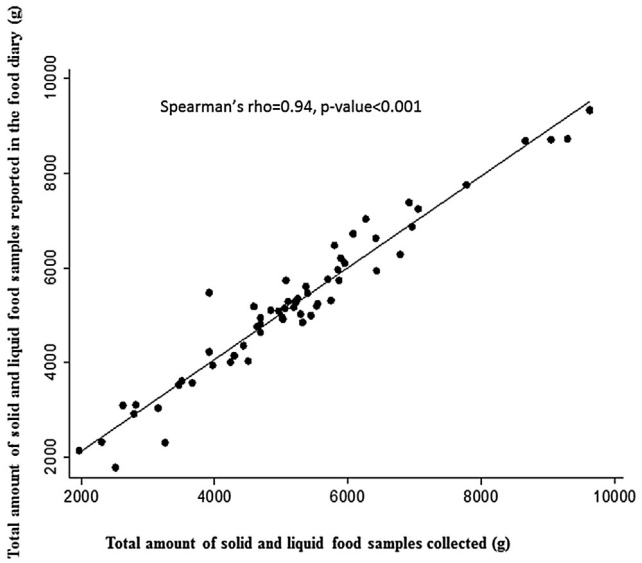


Fig. 3. Relationship between the total mass of solid and liquid foods recorded in the food diary and the mass collected in the duplicate diet study.

3.4. Indoor environment samples

The mass of collected floor dust significantly exceeded the mass of dust collected on the elevated surfaces in the same living rooms (Table 3). A positive correlation was found between the amounts of dust collected from the floor and the elevated surfaces and increasing living room size (in m^2) (floor dust, rho = 0.83, p-value <0.001; settled dust, rho = 0.76, p-value <0.001). As expected, the amount of dust from the vacuum cleaner bag was greater than the collected dust, as it represents accumulation of house dust for a longer period.

Regarding indoor air, 61 stationary air samples were collected for each family of compounds studied in this project while 15, 13, 16 and 17 personal air samples were collected for the analysis of PFASs, EBFRs, OPEs and PEs, respectively (Table 4). The average operating time of the personal pumps was 23 h. By comparison, the average sampling time of the stationary pumps in the participants' living rooms was 23.5 h, with 95% of the samples capturing 20–24 h sampling time. Finally, a total of 243 hand wipe samples were collected and a total of 3771 subsamples of dust, food/drinks, urine and serum were prepared.

3.5. A-TEAM rating form of sampling campaign

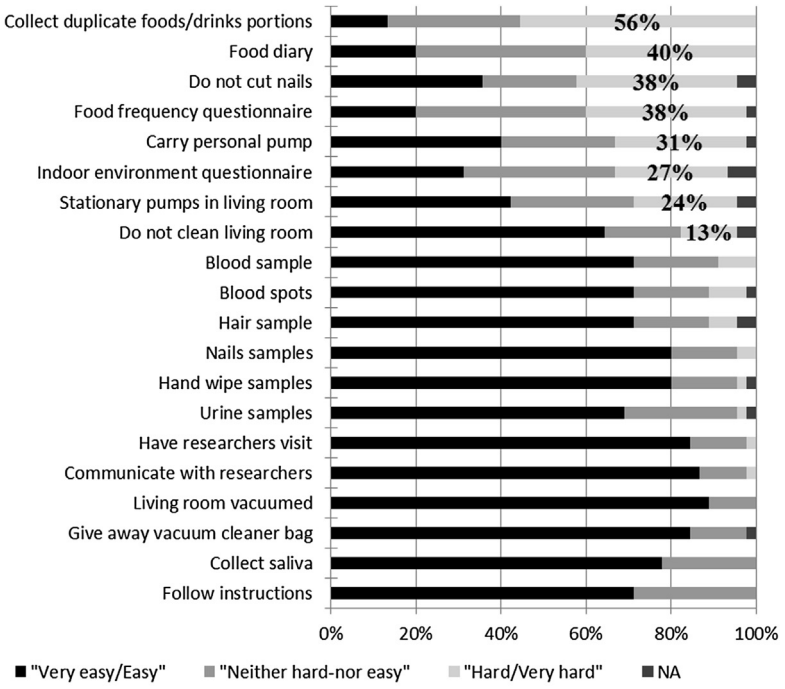
We received 45 completed forms from participants rating their experiences of the campaign (74% response rate). The gender-specific response rate was similar to the participation rate in the sampling campaign (76% women). Regarding their motivation to participate, most of the participants chose all three options given. Collecting duplicate portions of food and drinks was the task most frequently ranked as "hard/very hard" (56% of respondents) (Fig. 4). Recording the food diary and completing the FFQ was the next most difficult task. More than 60% of respondents stated the main reason to rate these tasks as "hard/very hard" was that they were "time consuming". Not cutting the fingernails was deemed "hard/very hard" for 38% of the respondents and was characterized as "complicated" by most respondents. Even though carrying the personal pump and having the stationary pump in the living room were not rated as "hard/very hard" by many respondents (31% and 24%, respectively); many reported that performing these tasks was "tiring" and "invasive" (82% and 96%, respectively). Nevertheless, 91% of the respondents were positive about participating in a similar project in the future.

4. Discussion

To our knowledge, this is one of the most comprehensive sampling campaigns conducted for the purpose of assessing human exposure to environmental contaminants from multiple exposure pathways. Based on the feedback from the participants, as well as the high sample collection rate, we consider this sampling campaign successful. The samples obtained provide a valuable resource for identification of the relative magnitude of different pathways of human exposure to PFASs, EBFRs, OPEs, and PEs. The main strength of our study design is the multi-pathway exposure analysis including multiple methods for assessment of exposure from each exposure pathway. Only a few studies exist that have performed this multipathway exposure analysis in such a

Table 4
Details of the indoor air, personal air and hand wipe samples collected.

Matrix	Contaminants			
	PFASs	EBFRs	OPEs	PEs
Indoor air				
Number of samples	61	61	61	
Number of field blanks	15	13	17	
Sorbent used	Env+ (0.2 g, 6 mL)	PUF & GFF	Env+ equipped with metal frits (0.2 g, 6 mL)	
Flow (L/min)	5	12	5	
Median Working time (hours, IQR)	24 (0.2)	24 (0.3)	24 (0.2)	
Personal air				
Number of samples	15	13	16	17
Number of field blanks	15	13	16	16
Sorbent used	Env+ (1 g, 25 mL)	PUF & GFF	Env+ (1 g, 25 mL)	Env+ (1 g, 25 mL)
Flow (L/min)	1	1	1	1
Median Working time (hours, IQR)	23.8 (1.2)	23.9 (0.5)	23.7 (0.5)	23.4 (0.7)
Hand wipes				
Number of samples	61	61	60	61
Number of field blanks	16	13	15	17
Sorbent	3 × 3 inches gauze pads	3 × 3 inches gauze pads	3 × 3 inches gauze pads	Glass wool (heated at 450 °C, 24 h)
Solvent	Isopropanol	Isopropanol	Isopropanol	Isopropanol
Container	125 mL PP bottle	60 mL amber glass bottle	125 mL PP bottle	125 mL PP bottle



Note: Frequencies of rating as "hard/very hard" are presented only for the tasks with more than 10% (≥5 participants) rating as "hard/very hard".

Fig. 4. All tasks performed during the A-TEAM sampling campaign and tasks rated as "hard/very hard" by participants (n = 45 responses).

comprehensive way [34,35]. Previous studies have assessed multiple exposure pathways by combining existing monitoring data [36] or have assessed single pathways [37]. This study provides a detailed snapshot of exposure (for one time point of 48 h exposure). Ideally this study should be repeated in the future to determine if exposure pathways have changed, although the resources necessary (12 PhD students and 3 postdoctoral researchers are employed in this study) are considerable.

Given the importance of the indoor environment for human exposure to consumer chemicals, indoor air, indoor dust and hand wipes were collected in our sampling campaign. Hand wipes were chosen as they can reflect dermal exposure from the surrounding environment after contact with contaminated surfaces, direct absorption from air and particle deposition [38]. In a study of office workers, positive relationships were reported between concentrations of PBDEs in indoor dust and hand wipes, as well as hand wipes and serum, suggesting that hand wipes can be an intermediate measure between external exposure and internal dose [30].

Regarding monitoring indoor air, we used both personal and static samplers in order to compare their efficiency as exposure indicators and to capture differences between indoor home exposure and that from outdoors or other microenvironments. Additionally, indoor dust was monitored by three collection approaches. We sampled dust from participant's vacuum cleaner bags which has the advantage of being easy to collect, and provides large mass of dust that represents an integrated and long-term measure of the whole house. However, factors related to the use of the vacuum cleaner can reduce the ability of this sample to reflect human exposure [10]. Hence, we decided to further collect a dust sample from the floor area and one from the elevated surfaces in the living room. Environmental chemicals are introduced in the food through environmental contamination of the food chain, or by transfer of chemicals from food packaging materials, during the food processing or storage and there are several approaches to assess the intake of contaminants through diet [39]. Even though there is a lack of knowledge regarding the validity of different dietary assessment methods for the intake of different chemicals, we have used three approaches to quantify daily dietary intake of the contaminants under study. FFQs are designed to assess habitual diet and have been used to calculate dietary intake of contaminants by combining the food intake data with food contamination data, mainly for persistent environmental pollutants [40,41]. The advantages of FFQ include the lower cost and less burden for the respondents and is recommended for large epidemiological studies [42]. Several duplicate diet studies have been conducted for the assessment of intake of several nutrients [43,44] and environmental contaminants from diet, such as dioxins, lead, arsenic, mercury, phthalates, PBDEs, PFOS and PFOA [6,45–49]. The advantage of the duplicate method to other food analysis methods along with the precise estimation of the content of chemicals in food is the incorporation of food cooking, storing and packaging at home, as well as composite meals. Additionally, the collection of duplicate food samples combined with a weighed food record provides additional data on consumption of individual foods. Such approaches are not subjected to recall bias, which is large disadvantage of the FFQ and are considered more precise methods. However, these methods are burdensome for participants and resource-intensive, do not reflect habitual diet and are suggested to be applicable in small-scale studies [42,50].

Serum and urine samples were collected in order to assess the body burden of the contaminants under study or their metabolites. By combining the different environmental and dietary monitoring approaches with body burden measurements, our aim is to identify those sampling methods that most accurately reflect human exposure to the studied consumer chemicals. Furthermore, the collection of blood spots, hair, fingernails and saliva as "non-invasive" samples provides added value to the project by facilitating assessment of the ability of such samples to represent human body

burdens adequately. Such evaluation has the potential to transform approaches to human biomonitoring and especially of young children, who are more susceptible to adverse effects related to exposure to chemicals. Nevertheless, we acknowledge that the samples collected provide a single point estimate of exposure, which may limit its ability to detect relationships between external and internal metrics of human exposure. However, associations between external and internal exposure would be possible in our project, provided that the measurements of external exposure (e.g. dust, air, hand wipes) are representative of exposure over an extended period, thus, the external exposure metric would be a major contributor to overall exposure. Another strength of our sampling campaign was the feedback obtained from participants. The decision to collect feedback was based on concerns about the burden placed on participants, given the many and demanding tasks asked over the 2-day sampling period. Importantly, the experience of the participants was positive, as reflected in their willingness to participate in a similar project in the future. Participants' views of the relative burden imposed by each task are extremely valuable in informing the design of similar future projects. Additionally, in this study, participants will be provided with feedback on concentrations of chemicals measured in their samples. The results from the rating form demonstrated that this was an important motivation for participating in the study. The potential for individual results to create anxiety among participants and mitigate negative impacts is acknowledged and we will carefully consider how and in what format we distribute and communicate results to participants [51].

A limitation of the present study is the non-representativeness of the study population that was recruited from the NIPH. Nevertheless, our intention was to evaluate a variety of approaches to measure external and internal exposure to consumer chemicals and the relationships between these, rather than to obtain representative exposure estimates of the general population.

In due course, data generated from analysis of the samples collected in this campaign will assist to further augment significantly understand of what constitutes best practice in human exposure assessment for our target contaminants and related chemicals. Importantly, our study reveals that, while 91% of participants were positive about future participation in a similar project, several burdensome aspects were highlighted. Mitigation of the impact of such tasks, combined with retention of positively-received components such as provision of feedback of results to individual participants, should form an important consideration in the design of future exposure assessment studies.

5. Conclusion

We conducted a comprehensive sampling campaign to study the multi-pathway human exposure to consumer chemicals and collected approximately 2000 samples of blood, blood spots, urine, nails, hair, saliva, foods, drinks, indoor dust, air and hand wipes from sixty one participants and their houses. The food collection and the completion of the food records were the most challenging tasks performed. Nevertheless, we believe that dietary intake data will be accurately estimated given the strong correlation between the total mass of food/drinks collected in the food bottles and recorded in the food records.

Important knowledge for us researchers included the assessment of the relative burden of each performed task, which may guide future studies to consider alternative ways to perform “hard” tasks, a positive participation experience can support re-participation, which might be important for longitudinal studies and the fact that providing feedback on the environmental monitoring results to the participants might motivate them for participation, which might be important for future similar biomonitoring studies.

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