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Estimating human exposure to perfluoroalkyl acids via solid food and drinks: Implementation and comparison of different dietary assessment methods

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ABSTRACT

Background: Diet is a major source of human exposure to hazardous environmental chemicals, including many perfluoroalkyl acids (PFAAs). Several assessment methods of dietary exposure to PFAAs have been used previously, but there is a lack of comparisons between methods.

Aim: To assess human exposure to PFAAs through diet by different methods and compare the results.

Methods: We studied the dietary exposure to PFAAs in 61 Norwegian adults (74% women, average age: 42 years) using three methods: i) by measuring daily PFAA intakes through a 1-day duplicate diet study (separately in solid and liquid foods), ii) by estimating intake after combining food contamination with food consumption data, as assessed by 2-day weighted food diaries and iii) by a Food Frequency Questionnaire (FFQ). We used existing food contamination data mainly from samples purchased in Norway and if not available, data from food purchased in other European countries were used. Duplicate diet samples (n = 122) were analysed by liquid chromatography coupled with tandem mass spectrometry (LC-MS/MS) to quantify 15 PFAAs (11 perfluoroalkyl carboxylates and 4 perfluoroalkyl sulfonates). Differences and correlations between measured and estimated intakes were assessed.

Results: The most abundant PFAAs in the duplicate diet samples were PFOA, PFOS and PFHxS and the median total intakes were 5.6 ng/day, 11 ng/day and 0.78 ng/day, respectively. PFOS and PFOA concentrations were higher in solid than liquid samples. PFOS was the main contributor to the contamination in the solid samples (median concentration 14 pg/g food), while it was PFOA in the liquid samples (median concentrations: 0.72 pg/g food). High intakes of fats, oils, and eggs were statistically significantly related to high intakes of PFOS and PFOA from solid foods. High intake of milk and consumption of alcoholic beverages, as well as food in paper container were related to high PFOA intakes from liquid foods.

PFOA intakes derived from food diary and FFQ were significantly higher than those derived from duplicate diet, but intakes of PFOS derived from food diary and FFQ were significantly lower than those derived from duplicate diet. We found a positive and statistically significant correlation between the PFOS intakes derived from duplicate diet with those using the food diary (rho = 0.26, p-value = 0.041), but not with the FFQ. Additionally, PFOA intakes derived by duplicate diet were significantly correlated with estimated intakes from liquid food derived from the food diary (rho = 0.34, p = 0.008) and estimated intakes from the FFQ (rho = 0.25, p-value = 0.055).

Conclusions: We provide evidence that a food diary or a FFQ-based method can provide comparable intake estimates to PFOS and PFOA intakes derived from a duplicate diet study. These less burdensome methods are valuable and reliable tools to assess dietary exposure to PFASs in human studies.

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1. Introduction

Food is a major source of human exposure to several persistent organic pollutants. Chemicals can be introduced to the food during food production, such as pesticides, hormones and antibiotics, during food processing, such as preservatives and packaging ingredients, or via the environment contaminate marine and/or agricultural food chains (Domingo and Nadal, 2017). Among such widespread food contaminants are the perfluoroalkyl acids (PFAAs). PFAAs are a group of highly fluorinated aliphatic substances that consist of a fully fluorinated carbon chain with different acidic functional groups attached (Buck et al., 2011). Due to their thermal and chemical stability and surface activity, PFAAs have been widely used in several consumer products and industrial application for decades (Paul et al., 2009).

Different PFAAs and their precursors enter the food and water through different pathways (Vestergren and Cousins, 2013). For example, PFOS and PFOA are regularly detected in foods through environmental contamination. It is expected that even though the production and use of PFOS and PFOA have been largely phased out in Europe and North America, they will continue entering food chains due to their long half-lives (Vestergren and Cousins, 2009). Precursors of major PFAAs, such as fluorotelomer alcohols and polyfluoroalkyl phosphate esters, are used in water- and grease-proof food packaging materials and can migrate to food (Gebbink et al., 2013; Trier et al., 2011).

Human exposure to per- and polyfluoroalkyl substances (PFASs) has been related to adverse health effects, even at low levels of exposure (Domingo, 2012). It is also of great concern that during pregnancy and in early childhood the vulnerable fetus and child is exposed to PFAS, as they are transferred through the placenta and through breastmilk (Papadopoulou et al., 2016b). Such early-life exposures have been linked to developmental toxicity (Johnson et al., 2014; Lau et al., 2007; Olsen et al., 2009), immunotoxicity (Mogensen et al., 2015) and several other health effects later in life.

Industry and regulatory authorities have made efforts to reduce the environmental release and further human exposure to PFOS and PFOA. Such efforts include; the 3 M phase-out of perfluorooctanyl (including PFOS and PFOA) chemistry in 2000–2002 (US EPA, 2000), the 2010/2015 PFOA Stewardship Program by eight participating companies (US EPA, 2015), the listing of PFOS and C_8-C_{14} PFCAs under the Candidate List of Substances of Very High Concern under the European chemicals regulation, REACH (ECHA, 2017) and the listing in Annex B (i.e. restricted use) of PFOS on the Stockholm Convention on Persistent Organic Pollutants (UNEP, 2015). National legislation has also been investigated, for example, in Norway, from 2014 it is not allowed to manufacture, import, export or sell consumer products containing PFOA, its salts and esters with content above 0.1% (Norwegian Environmental Agency, 2015). Despite these regulations, exposure to PFAAs continues and because of the phase-out of consumer products containing long-chain PFAAs, dietary exposure may have become relatively more important in recent years (Vestergren and Cousins, 2009). In several populations, diet has been identified as the major exposure pathway to PFAAs. Hence the continuation of food monitoring of PFAAs is essential for the assessment of the impact of environmental regulations and restrictions (Domingo and Nadal, 2017).

Human exposure to environmental contaminants can be assessed by different methods. In large population studies, the most frequently used method is to combine food contamination data, obtained through small or large scale food sampling and analysis (market basket studies, total diet studies) and food consumption data, obtained through food diaries or food frequency questionnaires (EFSA, 2012). In addition, the duplicate diet study, where participants are asked to collect an identical duplicate sample of their food as consumed, provides a snapshot of their dietary exposure to environmental contaminants at this time point, while it is a challenging and highly demanding task to perform (Papadopoulou et al., 2016a). Hence, different methods are associated with advantages and limitations. In addition, the use of different assessment methods might reduce comparability between and within populations, as they can represent acute, short-term or long-term exposure to contaminants. Comparisons of different dietary assessment methods for exposure to PFAA have not been performed and a few duplicate diet studies for dietary intakes of PFAA are available, leaving a large knowledge gap.

We measured the perfluoroalkyl acids (PFAAs) intakes of 61 Norwegian adults through a duplicate diet study. In addition, we estimated the dietary intakes of PFAAs for the same adults by using a food frequency questionnaire and a 2-days food diary, combined with a PFAA food contamination database. We compared and correlated the intakes derived from the three different methods using statistical methods.

2. Material and methods

2.1. Study population

This study is conducted within the A-TEAM project (Advanced Tools for Exposure Assessment and Biomonitoring). A-TEAM project's aim 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, emerging brominated flame retardants (EBFRs), organophosphate esters (OPEs) and phthalate esters (PEs). The sampling campaign conducted for the collection of several samples has been described in detail elsewhere (Papadopoulou et al., 2016a). In brief, 61 Norwegian men and women (74% women) were recruited and several samples relevant to both external and internal exposure were collected, including food samples, dust, air, hand wipes, blood and blood spots, saliva, hair, nails and urine, within a 2-day period. The average age of our participants 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, n = 57, 93%), were born in Norway (n = 44, 73%) and were non-smokers (n = 44, 73%). 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. Food collection and PFAS analysis of 1-day duplicate diet samples

During the duplicate diet study and after receiving detailed instructions, all participants collected a duplicate portion of all consumed foods and drinks, prepared as for consumption, over 2 consecutive weekdays. However, we have analysed PFAAs only in the duplicate diet samples collected in the 1st day. During collection of food and drink, they had to weigh and record all items that were prepared as duplicates in the food diary. Information on type, amount and time of consumption was reported for each consumed item, as well as 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). Participants were instructed to collect solid and liquid food samples in different 2 L polypropylene (PP) bottles for each day.

After collection, solid food samples were weighed, transferred into a food processor and blended for 2–5 min. Liquid samples were homogenized by hand shaking. After homogenization of each sample, 100 g subsamples of solid food and 100 mL subsamples of liquid food were stored in PP bottles and kept at −20 °C until analysis.

2.2.1. Sample preparation and analytical method

The method used for the sample preparation and instrumental