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Per- and polyfluoroalkyl substances (PFAS) in consumer products: Current knowledge and research gaps

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ABSTRACT

While several sources of per- and polyfluoroalkyl substances (PFAS) are known, their use in consumer household products is far less explored. The aim of this study was to provide comprehensive bottom-up analysis of the types and concentrations of PFAS reported in the literature over the past decade. A total of 52 studies revealed 107 PFAS belonging to 15 different categories in 1040 consumer products. The highest number of products tested were from the USA (n = 389) followed by the Czech Republic (n = 111). Mean PFAS concentrations were highest in household firefighting products, followed by textile finishing agents and household chemicals. The highest diversity of PFAS was reported in textiles (72 PFAS). Fluorotelomer alcohol (FTOH), polyfluoroalkyl phosphate esters (PAPs), perfluorocarboxylic acid (PFCA) and perfluorosulfonic acid (PFSA) are the classes of PFAS of high interest. Eight out of 52 studies used High-Resolution Mass Spectrometry techniques. Highlighted knowledge gaps included (i) the development of analytical methods for detecting a range of PFAS in consumer products, (ii) method validation and QA/QC approaches, (iii) application of suspect and non-target analysis, and (iv) an understanding of human exposure risk. This review highlights that the presence of PFAS in consumer products is of concern and remains underexplored.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a group of synthetic chemicals manufactured since the late 1940s and used in various industrial and consumer products to improve their physicochemical properties, such as enhanced resistance to heat, water, oil and stains (Buck et al., 2011). A recent comprehensive desktop study on PFAS in different applications estimated that > 1400 individual chemicals are used in > 200 different applications in industrial and household products (Gluge et al., 2020). Some identified use categories include coated cookware, water and stain-resistant clothes, food-handling materials, electroplating, fire-fighting foams, paints, additives in chemical manufacturing, and flame retardants (Gluge et al., 2020). PFAS used in their manufacture can be released into the environment, contributing to PFAS contamination. The global occurrence and distribution of PFAS in the environment, as well as human exposure, have been discussed in previous studies (Brumovsky et al., 2016; Cousins et al., 2022; Evich et al., 2022; Gebbink et al., 2017; Kurwadkar et al., 2022; Zhao et al., 2020).

Two principal manufacturing processes, electrochemical fluorination (ECF) and telomerisation are often used to produce PFAS (Buck et al., 2011). The ECF is an aggressive manufacturing process leading to many unwanted fluorinated by-products. In contrast, telomerisation produces homologous series of target compounds (Jackson and Mabury, 2013). Producing PFAS through these two approaches is costly; hence, PFAS are often used in products in which they are essential to gain the intended physicochemical properties. However, some studies have indicated that the non-intentional addition of PFAS to products can occur during the manufacturing process (Curtzwiler et al., 2021). Thus, knowing the exact details of PFAS being used in industrial and consumer products is particularly challenging. The confidential business information, insufficient product disclosure, and locations and quantities used in manufacturing have made it more complicated. Several studies have been conducted to analyse PFAS present in different products, for example, aqueous film forming foam (AFFF)(Barzen-Hanson et al., 2017), food contact material (Timshina et al., 2021), building material

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(Janousek et al., 2019), and hydraulic fluids (Zhu and Kannan, 2020a,b). PFAS produced through both ECF, and telomerisation have been reported in those studies.

A diverse range of materials and chemicals is used in consumer product manufacturing. It has been shown that these materials and chemicals pose significant long-term risks to human health such as DNA damage, reproductive/developmental toxicity and carcinogenicity (Akhavan et al., 2016; Knox et al., 2023; Li and Suh, 2019). More recently, studies have reported the presence of PFAS in various consumer products, with less explored production methods, which may contribute to human exposure. It has been shown that exposure to PFAS from these products can occur via several pathways, such as inhalation of dust and airborne particles, dermal contact with consumer products, household materials, and transfer from food contact materials (DeLuca et al., 2021). While human exposure to PFAS is primarily associated with ingestion and inhalation pathways, i.e., through diet, drinking water and indoor dust and air, pinpointing the most probable exposure pathway poses a unique challenge due to the complexity of the user pattern (De Silva et al., 2021). Exposure could be influenced by the nature of the products, the specific PFAS compound present and the frequency of product usage (DeLuca et al., 2021). The lack of comprehensive literature pertaining to PFAS-containing consumer products, makes it much more challenging to draw definitive conclusions. Substantial epidemiological evidence has reported an association between PFAS exposure and various detrimental health effects. These include carcinogenicity (Fenner, 2020), immunotoxicity (NTP, 2016), gene activation (Marques et al., 2022), and developmental toxicity (Fenton et al., 2021; Truong et al., 2022). However, analysing individual/group of consumer products to evaluate PFAS occurrence and potential human exposure is relatively new. Moreover, only a few selected PFAS were reported using targeted analytical methods. Thus, our current knowledge of PFAS composition in consumer products is limited. Consequently, we know very little about the possible release of PFAS into the environment and human exposure through consumer products (Balan

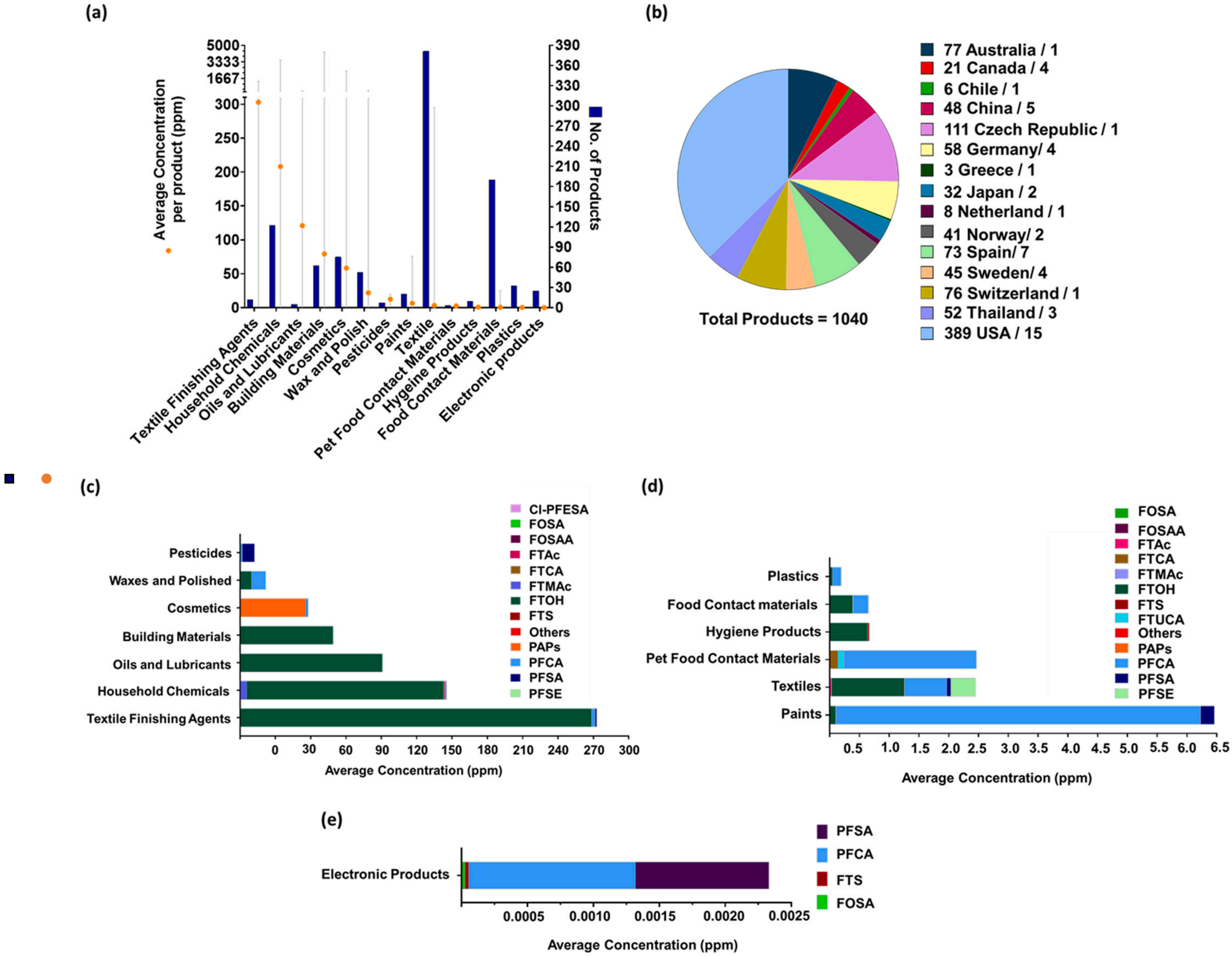

Fig. 1. Summary of the literatures found on PFAS-containing consumer products. (a) Number of PFAS containing consumer products (■) from the literature and the average concentration (ppm) of total PFAS content in the different PFAS categories (●). Error bars show the maximum and the minimum concentration of total PFAS in the product categories. Commercially available firefighting foams which could be used for domestic purposes were not considered for this graph but included in the supporting information. A study by Benotti et al. (2020) on food contact materials was also not included in this figure due to exceptionally high values reported compared to other studies in the category. (b) Number of consumer products analysed (n = 1040) for PFAS represented by different countries (Numbers prior to the country names represent number of products reported in that country and the numbers following the country name represent the number of studies reported). (c), (d) and (e) show the average sum of PFAS concentrations found in the different PFAS categories.

et al., 2021). Furthermore, while several top-down investigations have been published to predict PFAS in products, a bottom-up approach to provide a state of knowledge and assess the current research gap on PFAS in consumer products has not been reported to date.

Regulations for PFAS exposure in drinking and recreational water and food have been established for a limited number of PFAS (e.g. the US EPA's proposed National Primary Drinking Water Regulation (NPDWR) for six PFAS (EPA, 2022) and Australia's health-based guidance values for selected PFAS (NHMRC, 2019)). Guidelines for PFAS exposure through consumer products, however, has been overlooked due to a lack of comprehensive knowledge regarding the type and presence of PFAS in such products. This study aimed to provide a comprehensive analysis of PFAS reported in consumer products to understand the type, distribution, and concentrations of PFAS reported and inform better regulation and health protection. A secondary aim was to identify current knowledge gaps in evaluating human exposure through consumer products.

2. Methods

2.1. Literature review and search criteria

This review searched for literature reported PFAS in product types through direct experimental analysis. Literature was sourced from three reputable search engines: PubMed, Web of Science, and Scopus between August 2022 and April 2023. Search criteria were PFAS in consumer products, refined to within the last ten years and sorted by relevancy. All literature was available in English translations, and their references were further reviewed for relevancy to this review. The specific search terms used for the literature search are given in the supporting information (SI). Of these search terms, only fifteen product types (Fig. 1a, Table 1 and SI) were identified. Consumer products were determined to be items that the general public may come into contact with. Industrial chemicals were included where it was demonstrated that they would be used on products with a high likelihood of public exposure (i.e., clothing textiles). Studies reporting PFAS in AFFF samples not used by the general public were excluded. However, AFFFs commercially available to purchase for domestic purposes were included.

Fifty-two literature papers found were compiled into an Excel spreadsheet (see supporting information) under various categories to help sort and filter for data analysis. The categories were; paper cited (author and year published), product type, PFAS found, PFAS category, Concentrations (ppm), product types, usage status, target/non-target analysis, and country of testing. The product categories were defined based on the majority of product reported. Usage status was determined by whether the study had acquired the product directly from a shop or factory, and any product sourced after it had been purchased was considered used. Compiling the data into the spreadsheet started in August 2022 and was finalised at the start of April 2023 with several revisions. The Tables S1–S3 (are provided for data mining and have been submitted to NORMAN (Network of reference laboratories, research centres and related organisations for monitoring of emerging environmental substances) for suspect list exchange as a part of the NORMAN

Table 1

Number of literature, individual consumer products, PFAS found and mean concentration (ppm) of the total PFAS in different product categories.

	No. of papers	No. of products	No. of PFAS detected	Total PFAS concentration (ppm)			Reference
				Mean	Maximum	Minimum	
Textile finishing agents	1	12	13	302.92	1370.00	2.92	(Mumtaz et al., 2019)
Pet Food Contact Materials	1	3	10	2.42	5.63	0.21	(Timshina et al., 2021a,b)
Pesticides	2	7	2	12.36	19.20	3.92	(Fiedler et al., 2010; Lasee et al., 2022)
Building Materials	3	62	20	79.18	4300.28	0.0001	(Guo et al., 2009; Bečanová et al., 2016; Janousek et al., 2019)
Electronic Products	3	25	18	0.0023	0.03	0.0001	(Bečanová et al., 2016; Herzke et al., 2012; Meng et al., 2021)
Hygiene Products	3	9	9	0.66	2.90	0.0009	(Cécile and Hanssen, 2015; Guo et al., 2009; Supreeyasunthorn et al., 2016)
Cosmetics	4	75	50	58.21	2425.08	0.0020	(Fujii et al., 2013; Putz et al., 2022; Schultes et al., 2018; Whitehead et al., 2021)
Lubricants and Oils	4	5	33	121.05	396.00	0.0020	(Cécile and Hanssen, 2015; Fiedler et al., 2010; Yao et al., 2022; Zhu and Kannan, 2020b)
Paints	4	20	16	6.25	75.67	0.0020	(Bečanová et al., 2016; Favreau et al., 2017a,b; Herzke et al., 2012; Jia et al., 2021)
FireFighting Foams	5	39	21	488.51	11,031.30	0.11	(Benotti et al., 2020; Favreau et al., 2017a,b; Fiedler et al., 2010; Herzke et al., 2012; Weiner et al., 2013)
Plastics	6	32	22	0.19	2.61	0.0001	(Bečanová et al., 2016; Gomez et al., 2021; Liu et al., 2015, 2014; Llorca et al., 2014; Meng et al., 2021)
Household Chemicals	10	122	30	208.08	3490.60	0.0005	(Cécile and Hanssen, 2015; Daniel Borg, 2017; Favreau et al., 2017a,b; Fiedler et al., 2010; Guo et al., 2009; Herzke et al., 2012; Kotthoff et al., 2015; Liu et al., 2015, 2014)
Waxes and Polishes	10	52	24	21.85	423.40	0.0020	(Cécile and Hanssen, 2015; Daniel Borg, 2017; Fang et al., 2020; Favreau et al., 2017a,b; Ye et al., 2015; Gewurtz et al., 2009; Guo et al., 2009; Kotthoff et al., 2015; Liu et al., 2015, 2014)
Textile	21	381	72	2.48	295.20	0.0001	(Bečanová et al., 2016; Beesoon et al., 2012; Cécile and Hanssen, 2015; Gremmel et al., 2016; Guo et al., 2009; Janousek et al., 2019; Kotthoff et al., 2015; Liu et al., 2015, 2014; Meng et al., 2021; Muensterman et al., 2022; Rewerts et al., 2018; Schellenberger et al., 2022; Supreeyasunthorn et al., 2016; van der Veen et al., 2022; Wu et al., 2020; Xia et al., 2022; Zheng et al., 2020; Zhu and Kannan, 2020a; Rodgers et al., 2022)
Food Contact Materials	22	190	41	0.512	25.20	0.0001	(Benotti et al., 2020; Brenes et al., 2019; Cécile and Hanssen, 2015; Guo et al., 2009; Herzke et al., 2012; Kotthoff et al., 2015; Liu et al., 2015, 2014; Martinez-Moral and Tena, 2012; Moreta and Tena, 2014; Moreta and Tena, 2013; Poothong et al., 2012, 2013; Rewerts et al., 2018; Timshina et al., 2021; Yuan et al., 2016; Zabaleta et al., 2016, 2020, 2017; Zafeiraki et al., 2014

Joint Program of Activities (2023) on *analysis of PFAS using HRMS in consumer products*.

3. Results and discussion

3.1. PFAS containing products

Fifty-two studies investigating the presence of PFAS in 15 consumer product categories were identified (Table 1). Some studies examined multiple products belonging to a few different product categories in one study. Of these studies, 29% were from the USA, followed by Spain (13%) and China (10%) (Table S2). Germany, Sweden, and Canada were other significant contributors (i.e., four studies). The earliest study on PFAS-containing products was conducted by the U.S Environmental protection agency which commercial products were analysed for perfluorocarboxylic acid content (Guo et al., 2009). Since then, the number of publications in this field has shown an increasing trend, with a substantial rise from 2 in 2009 to 9 in 2022. Notably, 2021 and 2022 recorded the highest number of publications in this period, with 6 and 9 publications, respectively. Most of the studies were on food contact materials (FCM, n = 22 studies) and textiles (n = 21), followed by household chemicals (n = 10) and waxes (n = 10). Fig. 1 shows the number of consumer products from different product categories with a total of n = 1040 products analysed. Many used and unused textile products (n = 381) have been tested (including clothing, curtains, upholstery, sheets and blankets, carpets, and car textiles); of these, carpets and weather-resistant jackets were the ones most tested. Among the different FCM tested, microwave popcorn bags and fast food-related materials such as wrappings, food containers and straws are the most tested. Household chemicals included primarily carpet and fabric care products, impregnating agents, and general cleaning chemicals (i.e., dishwashing liquids, rinse aids, and rust removers), with carpet and fabric care products being the most common items investigated. A recent trend in investigating cosmetics has revealed 75 products tested across three countries (Japan, Sweden, and USA), including items such as foundations, mascaras, lip products, eye products, face products, concealers, and eyebrow products. Waxes and polishes used for various purposes have been tested as potential consumer products that contain PFAS. In addition, building materials that are used for general construction have also been analysed for PFAS. This included insulation materials, coatings, sealants, and foils. A few studies (5 studies) on commercially available household firefighting foam products (n = 39) which are normally kept for personal use were also found. While numerous potential applications of PFAS have been documented, the existing literature is from limited product categories. Therefore, it is imperative for future investigations to encompass a broader spectrum of household products (Gluge et al. 2020).

3.2. Types and concentrations of PFAS reported

The highest mean and maximum sum of total PFAS concentration were reported in foams from commercially available fire extinguishers (ranging from 0.1 to 11,031 ppm, mean 488 ppm, Table 1). Although AFFFs were not the focus of this study and studies reporting commercial use of AFFFs were excluded, these products were sold for domestic use. They were therefore included in the summary Table S1. However, the data was not included in Fig. 1 as our focus was on day-to-day consumer products. Twenty-one PFAS groups were detected in the firefighting foams, with PFOS, PFHxS, PFHpS and 6:2 FTS being the predominantly seen components in the mixtures, comparable to the AFFF used in professional firefighting (Barzen-Hanson et al., 2017). These findings are not surprising but highlight the need to investigate the phase out of PFAS in household fire extinguishers as well as the commercial formulations. The second highest concentrations were observed in textile finishing agents used in industrial textile preparations (mean 302 ppm), followed by household chemicals (mean 208 ppm). This implies that the PFAS found in textile products could be from the finishing agents used to achieve oil, water, and stain repellence (Mumtaz et al., 2019). However, the maximum concentration detected in textile (Rewerts et al., 2018) was substantially lower than that detected in textile finishing agents. Despite the comparatively lower mean PFAS concentration in household chemicals, the maximum concentration detected was more than double that of the textile finishing agents. Mainly, the PFAS amount detected in most of the finishing agents was significantly higher, and concentrations were > 1800 ppm in some products (Favreau et al., 2017). Building materials used for general-purpose constructions, such as coatings and insulation materials, also showed a significantly higher PFAS concentration. Notably, the highest PFAS concentration in building coating products (6:2 FTOH, 4300 ppm) only became second to commercially available firefighting foams (Janousek et al., 2019). This underscores the heightened risk of PFAS exposure for construction workers who routinely utilise these materials, emphasising the need for their inclusion in biomonitoring studies.

Fluorotelomer alcohols (FTOH) was the most frequently detected group in the tested product categories (data from commercially available firefighting foams was not considered), with the highest concentrations in textile finishing agents (mean 298 ppm), household chemicals (mean 167 ppm) and oils and lubricants (mean 121 ppm). Compared to the FTOH level reported, the concentration of all the other PFAS groups was substantially lower. However, FTOH concentration would not be a potential marker for PFAS fingerprinting as FTOH readily transforms to fluorotelomer carboxylic acids (Liu et al., 2007). Cosmetics, textiles, and FCM showed unique PFAS profiles. Polyfluoroalkyl phosphate esters (PAPs) were the most abundant group of PFAS (mean 55.5 ppm) found in cosmetic products, with 6:2 diPAP being the highest reported (2269 ppm). In contrast, PFCA dominated the PFAS profile (mean 2.22 ppm) in FMC, while FTOH, PFCA and perfluorosulfonamidoethanol (PFSE) were the dominant groups of PFAS in textiles. Of note, PFAS profiles in these three product categories were comparatively diverse and individual concentrations showed a significant variation. This implies a potential to have a similar PFAS exposure profile through these products. Some PFAS groups were only found in particular product categories indicating the unique functional use of those categories. For example, fluorotelomer methacrylate (FTMAc) was reported only in textile, cosmetic and impregnating agents. PFSE is another unique PFAS group detected in textiles, FCM and plastics.

The top product categories investigated (i.e., textiles, FCM, household chemicals and cosmetics) are all of concern due to frequent consumer use and a potentially higher chance of direct exposure. For example, textile products that are worn directly for extended periods, such as school uniforms, have a high potential to be a significant source of exposure (Xia et al., 2022). It has been estimated that the median potential dermal exposure through school uniforms was 1.03 ng/kg bw/day, which could be greater than the PFAS-containing occupational uniforms (Xia et al., 2022). It has been shown that there was a strong correlation between serum PFCA and PFSA levels and the consumption of fast food such as pizza and popcorn (Susmann et al., 2019). Supporting this, our summary clearly showed that microwavable popcorn bags were the most tested FCM, and PFCA is the common PFAS detected. A recent study showed that directly fluorinated containers, generally used in food packaging material, can be a significant source of PFAS exposure as short-chain PFCA migrate into the food from the directly fluorinated plastics with an estimated release of PFAS ranging from 0.77 to 2.68 ng/kg body weight per week (Whitehead and Peaslee, 2023).

4. Conclusions

Consumer products contain a wide range of PFAS (107 different PFAS) at various concentrations ranging from 0.26 ppt to 29,000 ppm. In addition to the well-known functional materials, such as wear and tear-resistant clothes and water and dirt-repellent chemicals, many different consumer products that have received lesser attention can contain a substantial amount of PFAS. For example, finishing agents and carpet care/cleaning products, which are frequently used in indoor environments, contain a high level of PFAS (6:2 FTOH). Cosmetics and personal care products showed a unique PFAS profile with diPAPs (e.g. 6:2 diPAP, 8:2 diPAP), which were minor compounds or absent in many other products. Different studies reported a significant variation among the PFAS categories and concentrations. For example, PFCA content in FCM reported by Benotti et al. (2020) was extremely high compared to all the other studies. The 6:2 PAP concentration reported from Canadian (0.0015 ppm), and Swedish (55.5 ppm) cosmetic foundations showed significant differences. PAPs are an analytically challenging class of PFAS to accurately quantify because of the hydrolysis of the phosphate ester bond and eventually transforming to PFCAs (Liu et al., 2007). Thus, sampling details, validation of analytical methods and QA/QC are of immense importance in reporting these types of PFAS. However, this information was not very clear in some of the literature. Of note, a trend of repetitive analysis of the same consumer products in different studies rather than focusing on the range of consumer products that might potentially contain PFAS was observed. Gluge et al. (2020) reported that ~200 consumer product categories are known to use PFAS during the manufacturing process, but a fraction of those categories has been tested so far. For example, there were no studies found on photographic materials which have been recognised as PFAS-containing products by analysis of manufacturing processes and patents.

Despite the complexity of the PFAS profile in consumer products, most of the studies have used target analysis with a selected suite of PFAS. Only eight studies (Rewerts et al., 2018; Yao et al., 2022) out of the 52 have used HRMS, which is necessary to detect a broad range of PFAS in a sample. Even though suspect screening and non-target analysis have been successfully applied to detect a wider range of PFAS from contaminated matrices (Barzen-Hanson et al., 2017; McCord and Strynar, 2019; McCord et al., 2022), these analytical approaches are not yet common in consumer product analysis. Among many different suspect lists compiled recently, such as PubChem and the NORMAN PFAS suspect lists, only one suspect list focusing on the PFAS from products is currently available (Gluge et al., 2020). It was clear from this work that there are knowledge gaps in (i) analytical methods for detecting a range of PFAS in consumer products (ii) method validation and QA/QC approaches, (iii) use of suspect and non-target analysis (iv) suspect list and non-target HRMS libraries with a focus on PFAS in consumer products. We consider this as a pilot study which can be expanded through collaborations and joint efforts to identify PFAS signatures in the different product categories to aid with a better understanding of PFAS sources and to be able to estimate individual household exposure in future. The summary table of this literature review will be available through NORMAN suspect list exchange. We welcome any inputs to expand the information in the summary table and, ultimately, aim to develop a comprehensive suspect list for screening consumer products for a wide range of PFAS.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data used for this study available in the supporting information.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.hazl.2023.100086.

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