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Review

Removal of perfluoroalkyl and polyfluoroalkyl substances from tap water by means of point-of-use treatment: A review

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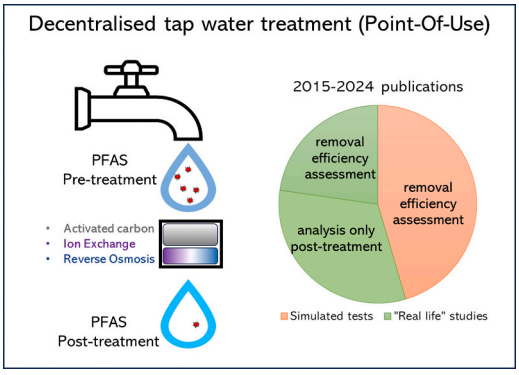
HIGHLIGHTS

- Point-of-Use treatments exploit technologies similar to those of centralized water treatments.
- A total of 22 peer-reviewed papers are present on PFAS removal by Point-of-Use treatments.
- Few works focused on evaluating POU efficiency in real conditions (occurrence studies).
- A limited set of PFAS is generally considered in evaluating the removal efficiency of the filters.
- AC and RO filters are mainly used and recommended, but correct handling should be addressed.

GRAPHICAL ABSTRACT



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ABSTRACT

Perfluoroalkyl and polyfluoroalkyl substances (PFAS) are widely used synthetic chemicals known for their environmental persistence and adverse health effects. For this reason, they have come under increasing scrutiny in drinking water, with several groundbreaking drinking water regulations adopted recently in the US and the EU. Nevertheless, conventional treatment processes often fail to remove PFAS effectively, raising concerns about drinking water quality and consumer health. More advanced treatment processes can remove PFAS with varying success from drinking water treatment plants. Using similar technology to that used in centralized PFAS treatment, many types of point-of-use/point-of-entry (POU/POE) water treatment devices are also commercially available.

Herein, an overview of the literature regarding POU/POE efficacy in the removal of PFAS from tap water was compiled and critically discussed. Generally, they employ treatment technologies like granular activated carbon, ion exchange, and reverse osmosis to remove PFAS contamination.

Despite their laboratory testing and often certification for removal of perfluorooctanoic and perfluorooctanesulfonic acid and other PFAS in tap water, in most cases their efficacy in actual use has yet to be well characterized. In particular, inconsistent testing and insufficient real-life studies complicate assessments of their long-term performance, especially against short-chain PFAS. Furthermore, improperly maintained activated carbon systems might even raise PFAS levels in purified water.

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Only a few peer-reviewed studies have measured PFAS levels at the tap after POU/POE treatment, with just five assessing removal efficiency in real-life scenarios. Limited to the findings described, not all filters were demonstrated to be effective, especially against short-chain PFAS.

Additionally, inconsistent testing methods that do not follow standard guidelines make it hard to compare filter results, and the long-term performance of these systems remains uncertain. More occurrence studies are essential to verify performance over time and understand exposure to these contaminants through water treated by household systems.

1. Introduction

Perfluoroalkyl and polyfluoroalkyl substances (PFAS) are a class of synthetically made chemicals. The general term "perfluoroalkyl(ated) substance," with the acronym PFAS, was the first to be defined and widely used to describe the broad class of highly fluorinated substances observed in the environment (Buck et al., 2011). Soon after, many authors also used the term "per- and polyfluorinated chemicals", with the acronym PFC which is also the acronym for perfluorocarbons and should be avoided. In 2011, the first clear structural definition of PFAS and recommendations on the names and acronyms for over 200 individual PFAS was published (Buck et al., 2011). Even if a universally accepted definition of PFAS is still lacking, this first harmonized PFAS definition was recently revised: PFAS refer to "fluorinated substances that contain at least one fully fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it), i.e., with a few noted exceptions, any chemical with at least a perfluorinated methyl group ($-CF_3$) or a perfluorinated methylene group ($-CF_2-$) is a PFAS" (Wang et al., 2021).

Since the 1940s, PFAS have been widely synthesized and utilized in various consumer, commercial, and industrial applications, e.g. used in cleaning products and lubricants, flame retardants, additives, pesticides, etc. (Gaines, 2023). In the last two decades, due to both occurrence studies and the documented adverse effect on living organisms, they have come under increasing scrutiny (Wang et al., 2021). "Legacy PFAS" – mainly long-chain PFAS with the most used and studied perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) – were gradually phased out. This has led to introducing and widely using emerging alternative PFAS, including shorter-chain perfluoroalkyl carboxylic acids (PFCAs) (fluorinated carbons $\leq$7), shorter-chain perfluoroalkane sulfonic acids (PFSAs) (fluorinated carbons $\leq$5) and other substances also containing heteroatoms, such as hexafluoropropylene oxide–dimer acid (HFPO-DA or more commonly GenX) (Li et al., 2020; Panieri et al., 2022). Very recently, the number of existing PFAS was reevaluated from several thousands to over seven million, following the revised PFAS definition (Schymanski et al., 2023).

PFAS are also known as "forever chemicals", due to their persistency (Wee and Aris, 2023). Indeed, they are chemically and thermally stable due to their multiple C—F bonds of high energy (531.5 kJ mol^{-1}) (Niu et al., 2016). They are also highly mobile and have been found even in remote regions far from sources of emissions (Panieri et al., 2022). In fact, a new planetary boundary for PFAS has been reportedly transgressed, with rainwater PFAS concentrations worldwide often above the strictest PFAS drinking water guideline values (Cousins et al., 2022). In addition, they are considered as amphiphilic substances, capable of bioaccumulation and biomagnification in biota through the contamination of food chains (Panieri et al., 2022). Still, this bioaccumulation is different from that of the classical persistent organic pollutants. If the latter, mainly lipophilic, preconcentrate in lipidic tissues (Khairy et al., 2021), PFAS highly accumulate in more hydrophilic matrices.

In humans, PFAS have been reported in multiple biological matrices such as blood and breast milk and represent a serious concern for human health, due to exposure of general population to contaminated food and water, and additional occupational exposure via air dust and derma. A number of adverse health effects have been correlated to PFAS exposure, yet they are still poorly understood and extremely challenging to interpret (Panieri et al., 2022; Wee and Aris, 2023). Some observed effects on animals and humans include endocrine dysfunction, alterations in the development, lipid metabolism, carcinogenicity, immunotoxicity and hepatotoxicity (Panieri et al., 2022). Based on effects on the immune system, a tolerable weekly intake for the sum of PFOS, PFOA, perfluorononanoic acid (PFNA) and perfluorohexanesulfonic acid (PFHxS) is 4.4 ng kg^{-1} body weight per week (Schrenk et al., 2020). Considering default assumptions for water consumption by an adult (2 L d^{-1}), body weight (60 kg) and an allocation factor of 20 % (the remaining 80 % coming from other sources than drinking water), a 'safe' drinking water limit of 3.8 ng L^{-1} would be derived for the sum of these four PFAS (Sadia et al., 2023). PFAS are now ubiquitous in the environment, including in drinking water sources and supplies (Vu and Wu, 2022; Wee and Aris, 2023). Drinking water consumption is one of the primary exposure pathways to PFAS for the general population, as highlighted in a recent review on the occurrence of PFAS in tap and bottled water worldwide (Teymoorian et al., 2023).

The most studied PFAS (PFOA and PFOS) have been detected at concentrations up to several μg L^{-1} in drinking water, while other PFAS in the order of ng L^{-1} (Wee and Aris, 2023). Different drinking water guideline values to protect the health of consumers have been proposed. At the European Union level, the revised EU Drinking Water Directive adopted in December 2020 set two group thresholds for PFAS: a limit for total PFAS at 0.5 μg L^{-1}, and the sum of 20 PFAS deemed to be of most concern at 0.1 μg L^{-1} (The European Parliament and the Council of the European Union, 2020). Member states should comply with the rules by the 12th of January 2026. Still, the method to measure "total PFAS" has not yet been defined. In April 2024, the US EPA announced a National Primary Drinking Water Regulation that establishes legally enforceable thresholds for six PFAS: individual maximum contaminant levels (MCLs) of 4 ng L^{-1} for PFOA and PFOS, of 10 ng L^{-1} for PFHxS, PFNA and GenX, and a hazard index approach for mixtures containing two or more of PFHxS, PFNA, GenX, and perfluorobutanesulfonic acid (PFBS) (EPA, 2024).

PFAS are difficult to treat due to their physicochemical properties, and the majority of traditional drinking water treatment processes are not able to remove PFAS. On the other hand, granular activated carbon (GAC), reverse osmosis (RO) and ion exchange (IEX) techniques have proven to be effective in removing long-chain PFAS. GAC is the most commonly used technology for PFAS control (Bertanza et al., 2020), while RO is particularly effective in removing both long- and short-chain PFAS (Wee and Aris, 2023). Treatment technologies for PFAS used on a large-scale application are generally the same as those used in smaller point-of use/point-of entry (POU/POE) applications.

POU/POE treatment is also called household water treatment or household water purification, or domestic filtration. POU treatment consists in a water treatment immediately prior to consumption, namely implemented just before its usage at a single water connection (tap). It differs from POE, which acts as a central treatment system of a whole building (U.S. EPA, 2006).

Considering the widespread contamination of drinking water sources by PFAS and the challenges in implementing a proper treatment in conventional full-scale treatment plants, POU/POE treatment can be a possible strategy to improve water quality (Vu and Wu, 2020; Wu et al., 2021). In fact, POU/POE systems have already been specifically implemented to reduce the concentrations of PFAS in tap waters impacted by contaminated groundwater (Mulhern et al., 2021). For example, POE

systems have been installed on wells used for potable purposes, that were contaminated with PFNA levels of ≥20 ng L^{-1} (New Jersey Drinking Water Quality Institute Health Effects Subcommittee, 2015). Still, there are relatively few peer-reviewed studies which assess the efficacy of POU/POE systems for PFAS removal in real-life scenarios (Table 1).

While several reviews have largely discussed the main technologies that are used in PFAS removal from drinking water, the focus was on centralized treatment (Crone et al., 2019; Lei et al., 2023). Only four of the first US studies on PFAS removal by POU/POE were briefly discussed in three previous reviews not focused on the topic (Lei et al., 2023; Reddy et al., 2022; Vu and Wu, 2022). In particular, Reddy et al. reviewed the performance of POU for EC removal, but mainly focused on describing existing technologies (Reddy et al., 2022). However, as far as the authors are aware, no previous scientific review article has extensively compiled and discussed the studies on PFAS removal by POU/POE treatment, especially in real-life scenarios. The present review fills the gap, discussing the technological trends in removing PFAS at the point-of-use, their current challenges (low concentration limits required, lack of information on the POU/POE systems in publications) and future perspectives.

2. POU/POE treatments: Overview

2.1. Treatment technologies

Since POU/POE systems are considered as an alternative to centralized water purification, their functioning principles are generally similar to those of large-scale water treatments.

The apparatus usually consists of one or more filter cartridges, with each filter characterized by a specific capacity. This is defined as the volume of feed solution that can be processed before the flow rate falls below a specified value (at constant pressure) or before the pressure exceeds a specified limit (at constant flow rate) (Ho, 2006). POE systems are connected to the water supply of a building and treat all the water that is distributed inside. POU systems on the other hand, can be directly connected to the water supply or non-fixed with manual refill. In the first case the system can generally be fixed (i) under the sink prior to the tap, (ii) as a countertop with a separate faucet, (iii) directly mounted on the faucet; in the second case portable pitchers are used. In both configurations, the cartridges can contain single sorption materials (especially when used in tandem) or a mixture of them, in order to adsorb different impurities from water. In fact, several principles of purification can be combined, such as mechanical filtration, adsorption on activated carbon (AC), IEX, RO and UV-light treatment (Manoleva et al., 2024; USEPA, 2024). Mechanical filtration aims at the removal of physical particulate, UV-light is mainly used for biological treatment, even though it may also be exploited to remove chemicals susceptible to UV110 degradation. Due to the lack of relevance for PFAS removal, mechanical filtration and UV-light will not be discussed (Pelayo et al., 2023). Regarding PFAS removal, especially in centralized purification, the most used technologies are by far GAC, IEX (in particular anion exchange) and RO. In POU, these technologies may be merged, by using pre- and post-filters, besides the "main" cartridge designed for the most effective PFAS removal. Indeed, since Natural Organic Matter (NOM) is able to interfere with PFAS removal (Dixit et al., 2021; Yu et al., 2012; Zhang et al., 2019), the use of pre-filters can help in its removal, enhancing the overall efficacy. Moreover, pre-filters are particularly necessary when RO cartridges are used, to increase their lifetime and avoid overpressure issues.

2.1.1. Granular activated carbon

GAC is a microporous adsorbent with a large surface area, whose structure is based on carbon atoms ordered in parallel stacks of hexagonal layers, extensively cross-linked and tetrahedrally bonded. In addition to that, several heteroatoms, such as oxygen, hydrogen and nitrogen can be found in the carbon matrix, both interspersed within the fullerene-like carbon network and/or in the form of surface functional groups (Karanfil et al., 1999; Zhi and Liu, 2015). Thus, GAC structure and properties generate several possible interactions with organic chemicals. Mainly, adsorption is controlled by physical interactions

Table 1
occurrence studies that included measurement of PFAS in tap water samples after POU/POE treatment.

Country	Knowledge on types of filtration used (AC, IEX, RO)	Number of PFAS studied	Knowledge on 'filter age'	Sample extraction	Analytical instrument	LODs or LOQs (ng L^{-1})	References
Thailand	NR	11	NR	SPE (Presep-C Agri)	NR	NR	Tabtong et al. (2015)
US	Incomplete (GAC)	7	NR	direct analysis	LC-MS/MS	3–6 (LODs)	Scher et al. (2018)
China	NR	6	NR	SPE (WAX)	LC-MS/MS	NR	Ao et al. (2019)
US	Incomplete (AC and RO)	39	NR	Direct analysis	LC-MS/MS	0.12–23.3 (LOQs)	Bradley et al. (2020)
Thailand	NR	33	NR	SPE (HLB-WAX in tandem)	LC-MS/MS	0.03–0.13 (LOQs)[a]	Guardian et al. (2020)
US	Incomplete	34	NR	Direct analysis	LC–MS/MS	Reporting Limit 1.9–50.5 (RL)	Bradley et al. (2022)
Spain	Incomplete (AC and RO)	35	NR	SPE (a combination of mixed-mode Strata-X cartridge and plus Hypersil GOLD C18)	LC–MS/MS	1–50 (LOQs)	Cserbik et al. (2024)
US	Incomplete (IEX and GAC)	Non-targeted	NR	SPE (WAX)	LC-HRMS	NR	McCord et al. (2020)
US	Yes (AC and RO, also combined)	11 or 6	Yes	SPE (WAX)	LC-MS/MS	<0.01–2.68 (LOQs)	Herkert et al. (2020)
China	Yes (AC and RO combined)	32	Yes	SPE (WAX or HLB)	LC-MS/MS	10–240 (MDLs)	He et al. (2022)
US	Yes (AC block)	24 to 47	Yes	SPE (Strata™-X-AW)	LC–MS/MS	0.5–6.2 (MRLs)	Mulhern et al. (2021)
Spain	Incomplete (AC and RO)	35	NR	SPE (a combination of mixed-mode Strata-X cartridge and plus Hypersil GOLD C18)	LC–MS/MS or LC–HRMS	1–50 (LOQs)	Cserbik et al. (2023)

AC: activated carbon; LOQ: limit of quantification; MDL: method detection limit; MRL: method reporting limit; NR: not reported; RO: reverse osmosis; SPE: solid-phase extraction; WAX: weak anion exchange; HLB: Hydrophilic lipophilic balance.

[a] For 15 detected PFAS only.

(given by size exclusion and microporosity effect) and chemical interactions, depending on the chemical nature of the surface, the adsorbate, and the solvent (Karanfil et al., 1999). In the adsorption of PFAS, both types of interactions are involved. Hydrophobic interaction is reported as the main sorption mechanism, implying a higher removal of long-chain PFAS by GAC (Du et al., 2014; Wang et al., 2019; Zhang et al., 2019). Besides, the functional groups may provide additional features, such as surface charge, which can confer acidic-basic properties to GAC, consequently leading to electrostatic interactions between GAC surface and the negatively-charged PFAS (Cantoni et al., 2021). Indeed, modified carbons with basic surface properties have shown a higher ability to remove PFAS (Zhi and Liu, 2016). On the other hand, the physical mechanism of size exclusion can be tricky for higher molecular weight compounds or PFAS micelles, since this phenomenon may cause a reduction in the sorption ability when smaller micropores become inaccessible (Pauletto and Bandosz, 2022; Siriwardena et al., 2019). Finally, GAC with a larger available surface area (higher porosity/smaller particles) was shown to be more effective in interacting with short-chain PFAS (Cantoni et al., 2021). Still, there is a consensus in highlighting the low efficacy of GAC in removing these more hydrophilic PFAS (Li et al., 2020; Zhang et al., 2019).

2.1.2. Reverse osmosis

RO is a filtration mechanism which exploits the different permeability of a porous membrane with respect to solvent and solutes. In particular, in contrast with osmosis, it is a process during which an outer pressure is applied to force the solvent through a membrane (permeable to the solvent) and rejects the solute, thus creating the permeate and retentate streams, respectively (Mastropietro et al., 2021; Spohn, 2000). The mechanism involved in RO is basically the same as in other membrane-based systems, such as micro-, ultra- and nano-filtration, RO being the most extreme among them. In these approaches, pore size goes from 0.1 to 10 μm for microfiltration, enabling the retention of large colloids and bacteria, up to 0.1 nm for RO, in which also monovalent ions are mostly retained (Yang et al., 2019). The main advantages of membrane technology over other purification methods are high removal capacity, modularity and flexibility. Still, higher costs and operational requirements (pressures up to 100 bars), as well as susceptibility to fouling, constitute the major drawbacks. The rejection ability of RO processes depends on size exclusion and solute-membrane interaction (Mastropietro et al., 2021). This can be in turn differentiated in electrostatic repulsion between charged solutes and membrane surface and other chemical interactions (hydrophobic, hydrogen bonding and dipole interactions). RO represents a promising approach for PFAS removal, even for shorter-chain compounds (Tow et al., 2021). If size exclusion seems to be the main mechanism in the rejection of PFAS by RO membranes, electrostatic repulsion also plays an important role. Indeed, the most used RO membranes are made of polyamide, normally negatively charged under ambient pH (Liu et al., 2022).

2.1.3. Ion exchange resins

IEX resins are polymeric materials characterized by the presence of charged functional groups, which allow the process of reversible exchange of positive or negative counter ions from the surface of the resin to its surrounding aqueous matrix (Dixit et al., 2021). The efficacy in removing charged contaminants depends on their affinity for the resin surface, compared to those of other ions which can be present in the aqueous matrix. In the case of PFAS, anion exchange resins proved to be the most effective, due to the negatively charged moiety of most perfluorinated compounds. The polymeric backbone of these materials is generally acrylic or styrenic, while the functional groups can be tertiary or quaternary ammonium groups (weak and strong IEX resins, respectively) (Gao et al., 2017; Maimaiti et al., 2018; Schuricht et al., 2017). Both ionic and non-ionic resins have been tested for PFAS removal, with the ionic ones showing better performances (Du et al., 2014).

Moreover, when studying several perfluorinated compounds, the negative atomic charge showed better correlation with PFAS removal, than the hydrophobicity (Park et al., 2020). Still, the different affinity shown for different polymer bulks revealed that hydrophobic interaction may increase the efficacy of certain IEX resins in removing PFAS (Zhang et al., 2019).

The possible exploitation of both hydrophobic and electrostatic interactions represents the main advantage of IEX resins, which also demonstrated to be more effective towards eliminating emerging short-chain PFAS than carbon-based adsorption materials (Dixit et al., 2021).

Another important advantage is represented by their relatively easy recovery, obtained by using salt-containing solutions. Negatively charged counter ions with high charge density are in fact able to replace the adsorbed PFAS, in some cases in combination with organic solvents (Dixit et al., 2020; Gao et al., 2017).

2.2. Performance evaluations and certification

To address the need for consensus methods for evaluating the effectiveness of POU/POE technologies for treating PFAS contamination of drinking water, POU/POE systems can be certified by independent third-party accreditation organizations using American National Standards Institute (ANSI) standards to verify contaminant removal claims. In 2016, a standard test method and protocol for the reduction of PFOA and PFOS (NSF P473-Drinking Water Treatment Units – PFOA and PFOS), was developed by National Science Foundation (NSF) International (NSF, 2022). According to the test protocol, spiked water concentrations of PFOA (500 ng L^{-1}) and PFOS (1000 ng L^{-1}) needed to be reduced to a sum below the USEPA health advisory levels of 70 ng L^{-1}. NSF P473 was then retired and incorporated into two existing water treatment standards, NSF/ANSI Standard 53 (Drinking Water Treatment Units – Health Effects for the reduction of PFOA and PFOS with granular activated carbon filtration or anion exchange media systems) and NSF/ANSI Standard 58 (Reverse Osmosis Drinking Water Treatment Systems for the reduction of PFOA and PFOS with reverse osmosis systems) (Racz and Kempisty, 2021). The most recent editions (2022 versions) revised the requirements for PFOA and PFOS based on current health advisory levels and expanded the chemical reduction claims to include five new PFAS: PFHxS, PFNA, perfluoroheptanoic acid (PFHpA), PFBS, and perfluorodecanoic acid (PFDA) (Kelechava, 2023).

The NSF/ANSI testing procedures utilize a “challenge” influent water to test POU/POE efficacy. This water meets or exceeds 95 % of the concentration levels previously found in drinking water based on a dataset compiled from the third Unregulated Contaminant Monitoring Rule US campaign (UCMR 3) and an additional dataset from the Environmental Working Group (EWG). The challenge water contains a mixture of seven PFAS compounds with a total concentration of 2160 ng L^{-1}, made up of PFOA (500 ng L^{-1}), PFOS (1000 ng L^{-1}), PFHxS (300 ng L^{-1}), PFNA (50 ng L^{-1}), PFHpA (40 ng L^{-1}), PFBS (260 ng L^{-1}), and PFDA (10 ng L^{-1}) (Andrew, 2023a, 2023b). The reduction of PFAS is considered acceptable by the protocol in 2 cases: the initial “total PFAS” concentration is reduced by at least 99 % (effluent concentration of 20 ng L^{-1}); the concentration of the individual PFAS compounds reach their respective health advisory concentrations in tap water. These values are 20 ng L^{-1} for PFOA and PFOS combined, 20 ng L^{-1} for PFHpA and PFHxS individually, and 6 ng L^{-1} for PFNA (Andrew, 2023a, 2023b). Two PFAS compounds, PFBS and PFDA, are excluded from the individual contaminant reductions because their occurrence levels were found to be less than their health advisory level at that time (Andrew, 2023a, 2023b).

Present POU/POE treatment systems may be used to reduce PFAS concentrations, but they may not ultimately be acceptable for compliance of drinking water systems in the US, based on the recent National Primary Drinking Water Regulation. Indeed, current NSF/ANSI certification standards do not yet indicate that a POU/POE filter will remove PFAS down to their MCLs. However, POU/POE treatment is reasonably anticipated by the EPA to become an attractive compliance option for

small water system consumers and private well owners in the future, once NSF/ANSI or other independent third-party certification organizations have developed a new certification standard to demonstrate the ability of the devices to meet the EPA's MCLs (EPA, 2024b).

Data on PFAS removal are sometimes presented by companies providing POU/POE systems. Still, often, the claim to be able to remove PFAS are not supported by rigorous tests, or the results are not presented/justified. Sometimes, untested PFAS are claimed to be removed just based on tests on other compounds. For example, on a company website, GenX was hypothesized to be removed only because PFOA removal was verified. These claims must be considered with care, since GenX has been found to be less easily removable than PFOA (Herkert et al., 2020). Others indicate the removal percentages for many PFAS by their POU/POE systems according to outside laboratories. Still, it is advisable to check what lab test were actually conducted and verify that removal claims were not extended to PFAS other than those actually tested.

3. Studies on PFAS removal by POU/POE

3.1. A recent research topic

In general, literature is lacking peer-reviewed studies regarding PFAS removal by POU/POE systems. Fig. 1 summarizes all papers on the topic, by distinguishing the research conducted in the laboratory and that regarding actual use of the POU/POE in real life. The bibliographic research methodology can be found in the supporting material.

To the best of the authors knowledge, the first two works appeared in 2015, and a total of only 22 peer-reviewed papers has been published up to now (2024). The studies can be distinguished in two main types:

- Lab-studies- tests performed in controlled conditions in the lab or in simulated situations in the field ("stress situations").
- Occurrence studies- tests performed in real-life situations of POU/POE usage.

Even if relatively few studies are currently present in the literature, the number is expected to rapidly grow, given the increasing interest in these substances. The growing awareness of the presence of PFAS all over the world (Ackerman Grunfeld et al., 2024) creates concerns about their concentrations in drinking water. In parallel, there is a rapidly growing use of POU/POE systems in both developed and developing countries, especially where centralized treatment is not applied.

Regarding PFAS in general, the US have published the highest total number of articles on these substances (Klingelhöfer et al., 2024). Currently, North America also leads the world market for POU/POE water treatment systems (Reports and Data, 2023). In addition centralized water treatment is absent in many regions (> 25 million private water users) (Dieter et al., 2018). This, alongside with the awareness of PFAS contamination and current financial incentives, could explain why nearly half of the studies (10 out of 22) on PFAS removal by POU/POE found so far in the literature are from the United States. Recently, Chinese researchers are focusing more and more on studies regarding PFAS in general (Klingelhöfer et al., 2024), including toxicity studies, occurrence studies and remediation technologies, due to the rapidly increasing contamination by PFAS in China in the last ten years (Baluyot et al., 2021). This explains why China is the country with the second highest number of publications (3) on POU/POE systems for PFAS removal. The global POU/POE water treatment systems market size is predicted to experience more than a fivefold increase in the next ten years, especially in Asia-Pacific (Reports and Data, 2023). With increasing use of POU/POE systems and stricter regulations on PFAS in drinking water, more publications regarding PFAS removal by POU/POE systems can be expected in the future.

3.2. Lab studies

Lab studies on the topic include both non-peer review reports and peer-reviewed articles. The earliest POU/POE study that can be found in the literature was an evaluation of the efficiency and effectiveness of POU treatment systems in a non-peer reviewed report of the Minnesota Department of Health (Olsen and Paulson, 2008). Fourteen filters were lab-tested: six AC block devices and eight RO systems operated with an AC post filter. At that time, there was no NSF standard for evaluating PFAS reduction. Lab tests were performed with high initial PFAS concentrations of 3000–10,000 ng L^{-1}, by assessing PFOA, PFOS and PFBA only. Still, the tests were quite vigorous: for example, the tested AC filters were operated to 150 % of their manufacturers rated capacity and RO systems for 90 continuous days. POU systems that passed initial lab testing (11/14) were field-tested using water from municipal wells contaminated by PFAS and were found to remove PFAS in field tests to below the employed detection limits (50 ng L^{-1}) (Olsen and Paulson, 2008). Later, in 2016, the Minnesota Department of Health also tested up to 100 % of its capacity a faucet-mounted carbon filter for its effectiveness in removing seven PFAS. In their test, six of the seven PFAS were reduced down to non-detect levels (MDH, 2017). In a work funded by the Water Quality Research Foundation, three POU RO membranes and three AC filters were tested for the removal of 3 PFAS (PFOS, PFBS,

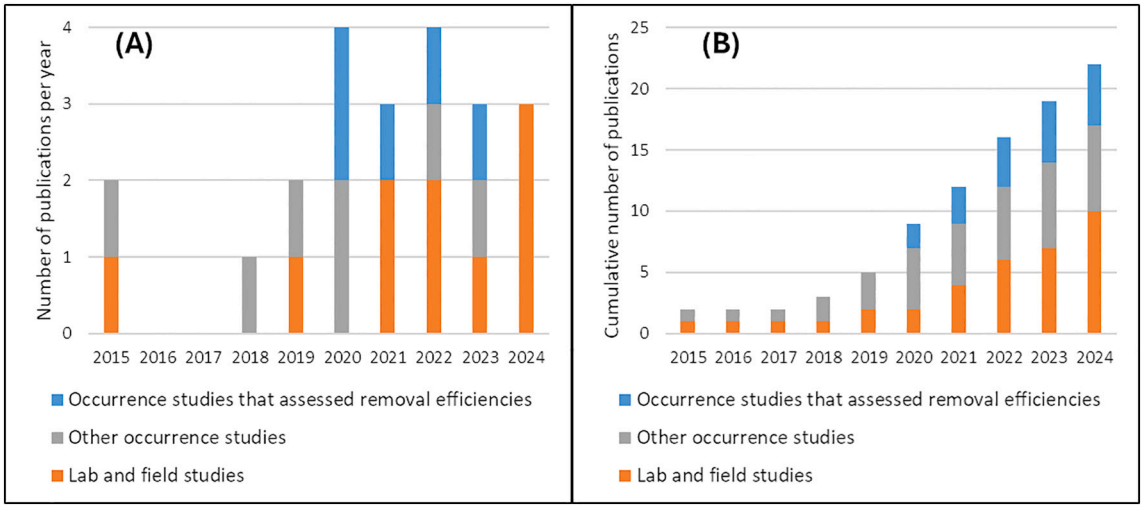

Fig. 1. Number of scientific publications on the removal of PFAS by POU/POE in tap water: (A) per year; (B) cumulative.

PFHxS) at two different concentrations (1 μg L^{-1} and 10 μg L^{-1}) in spiked water (Zhou, 2022). AC filters were tested to 200 % of their designed treatment capacities, and RO membranes with recovery of 19.3 % (percentage of feed water converted to produced water) were tested for three weeks. The tested PFAS were effectively removed by both RO membranes and AC filters, with average removal efficiencies >90 % in all POU devices. More recently, the Environmental Working Group (EWG) tested 10 pitcher/countertop water filter systems for their ability to remove a larger amount of PFAS from tap water (Lacey et al., 2023). In principle, 25 individual PFAS were considered, but only 9 were detected in two tap water samples prior to filtration and no spike was added: long-chain PFAS (PFOA, PFOS, 6:2-fluorotelomersulfonic acid (6,2-FTS)) as well as short-chain PFAS (PFBA, PFBS, PFHxS, perfluorohexanoic acid (PFHxA), perfluoropentanoic acid (PFPeA), perfluoro-4-oxapentanoic acid (PF4OPeA)). In the experiment, 10 gal (approximately 38 L) of non-spiked unfiltered tap water were passed through each water filter, regardless of its capacity, and after the process one single sample was collected for each filter. Pitcher filters varied widely in their ability to remove PFAS, with elimination percentages in the tested conditions ranging from 22 % to 100 % depending on the brands.

Very recently, seventeen POU systems (faucet mounted, refrigerator, plumbed-in, which includes filters that can be connected under the sink or placed on the countertop near the sink, and standalone) were tested for PFHxS, PFHpA, PFOA, PFOS, PFNA, PFDA removal using contaminated groundwater (total concentration of 122.6 ng L^{-1}) (MassDEP and Umass Amherst, 2023). Three of these were NSF/ANSI Standard 53 certified. Samples were collected at 100 % and 200 % of the manufacturer-rated throughput volume capacity. On the other hand, twelve non-certified filters were not able to produce water with the sum of PFAS below 20 ng L^{-1}, at 100 % of the manufacturer recommended volume capacity.

The peer-reviewed publications include nine treatability studies, namely performed at the laboratory and generally (but not always) by setting known PFAS concentrations as input, and one field test study, regarding the treatment of contaminated water by a POE in simulated conditions. Often, such tests involve passing high volumes of water in a short time frame, thus potentially compromising their significance compared to real-life less intensive use of POUs. In the first peer-reviewed publication in 2015, Anumol et al. investigated the removal of PFOA and PFOS (within a group of sixteen trace organic compounds), by 3 commercially available pitcher POU devices and 2 refrigerator-fitted POU devices, all exploiting AC (Anumol et al., 2015). The tests were performed according to NSF/ANSI Standard 53 guidelines and results showed that the refrigerator filters were more efficient for the removal of the two PFAS. Among the pitcher filters, the presence of anion exchange resins in specific filters was thought to improve the removal efficiency. Patterson et al. tested 3 commercially available RO treatment systems and 2 GAC media at various flow rates, with initial PFAS concentrations of 200–1600 ng L^{-1} (worst-case concentrations compiled from historic records (2013–2016) of Widefield aquifer region water samples). Samples were collected every half hour over an 8 h period for GAC, and every 2–3 times per day for 1 week for RO. Effective removal of six legacy perfluoroalkyl acids (PFOS, PFOA, PFHpA, PFHxS, PFBS, and PFNA) was demonstrated (Patterson et al., 2019).

Iwabuchi and Sato tested four different models of pitcher-type water filters from four manufacturers – two with a carbon, ceramic, and hollow fiber membrane design; and two with an AC and IEX design (Iwabuchi and Sato, 2021). All tests were performed by loading water with initial concentrations of 50 ng L^{-1} of each of six considered compounds (PFOA, PFOS, PFHxA, PFDA, perfluorododecanoic acid (PFDoA), and PFHxS). One liter of the test water was applied to the water filters 200 times, with the filtrate analysed after 10 L, 100 L, and 200 L had been passed through the filters. As could be expected, for each system, filtration effectiveness decreased with prolonged use. Removal efficiency varied by carbon-chain length (i.e., more efficient removal was observed for longer-carbon-chain PFAS) and the PFAS functional group (PFOS >PFOA, and PFHxS >PFHxA). These results substantially agree with studies regarding central purification systems exploiting carbonaceous materials, which highlighted the complexity of the interactions between the material and the different compounds, but generally conveying a more efficient removal of the more hydrophobic PFAS (Cantoni et al., 2021; Ochoa-Herrera and Sierra-Alvarez, 2008; Westreich et al., 2018).

Breakthrough plots of individual and total-PFAS as well as optical parameters were studied in column tests (pilot and "rapid small-scale column test") of granular biochar designed around a household treatment unit (Aung et al., 2022). Test waters were spiked with 11 short-chain PFAS to obtain final concentrations of individual PFAS of 400 to 500 ng L^{-1} each (Aung et al., 2022). The results showed that ultraviolet absorbance at 254 nm (UV254) and total fluorescence (TF) could be used as surrogates to predict the removal of the sum of individual PFAS and for 9 of the 11 PFAS on an individual basis, by carbon adsorption. Still, UV254 and TF may be influenced not only by the PFAS concentration but also by the overall water chemical composition. Thus, the results obtained by applying these methods must be taken with caution.

Zambianchi et al. developed a polysulfone graphite oxide hollow fiber membrane (PSU-GO HFs) to synergize ultrafiltration and adsorption mechanisms for enhanced drinking water treatment at the POU. The removal efficiency of PSU-GO HFs was better than that of PSU with GO or that of commercial GAC for nearly all the tested PFAS (14 of different molecular size (C3-C13) and end substitution) (Zambianchi et al., 2022). It is important to note that while the design was clearly destined for POU treatment, these tests were conducted in lab-scale modules and no preliminary tests on POU real scale filters have yet been conducted for PFAS. Khaliha et al. later used scraps obtained as waste of the industrial production of PSU-HF and PSU–GO-HF, converted into PSU and PSU-GO granular materials (Khaliha et al., 2024). These were tested alongside GAC in small prototype cartridges for the same PFAS with similar results to those of Zambianchi et al. In addition, testing of the three materials in standard sized POU commercial cartridges, for PFOA only, resulted in better absorption capacity of the PSU-GO granular materials (Khaliha et al., 2024). While nanomaterials are promising to improve POU/POE treatment, some issues remain concerning the cost, risk of release of nanomaterials into the treated water and public acceptance compared to traditional POU/POE treatment options (Kidd et al., 2021, 2020; Prathna et al., 2018; Westerhoff et al., 2016).

Liang et al. analysed the waters from 10 taps in China for 7 ultrashort-chain and short-chain PFAS (C1-C4). For one of these tap waters, a sample was also analysed after RO treatment in the research facility with no further information. All 10 tap water samples contained trifluoroacetic acid (C1), while it was not detected in the RO-treated sample (Liang et al., 2023).

Recently, five different brands of pitchers were assessed for the removal of 75 PFAS from two different water sources in Canada (Teymoorian et al., 2024). Roughly 50 PFAS were detected in the source waters prior to the lab tests. Filtered water was analysed after 20, 40, 80 and 160 L had been filtered, which only covered the lifetime of two of the five pitchers. The two NSF or WQA certified filters demonstrated the highest PFAS removal performances (close to 100 %). For the other three filters, a removal between 38 and 60 % was observed, with better efficiencies for PFAS with increasing chain length, and for PFSAs compared to PFCAs. The best pitcher was also tested in some homes, and the results were similar to the laboratory tests.

Cappelli et al. also used a new commercial pitcher containing activated carbon from coconut shells to filter 8 different Belgian tap waters for 7 ultrashort-chain and short-chain PFAS (C1-C4). The commercial filter significantly reduced the median concentrations of the studied compounds in tap water (Cappelli et al., 2024). Nevertheless, as rightly noted by the authors of the study, the test only assessed the 'best removal with the first usage' and provided no longitudinal information (Cappelli et al., 2024).

A POE adsorption system with 5 filter cartridges in series (with

different media) was tested for removal of 30 PFAS (perfluoroalkyl acids and some of their precursors) in groundwater contaminated with aqueous film-forming foam (Natarajan et al., 2021). Eleven PFAS were detected in the groundwater (10–680 ng L^{-1}). Once again, the long-chain PFAS were more effectively removed than short-chain PFAS, and PFCAs broke through before PFSAs of the same chain length. The studied POE filters showed 5 months of effective removal for all the PFAS measured, and over 7 months for most of them, except shorter-chain PFCAs.

The described laboratory studies highlight a very strong tendency of filters to allow increased breakthrough (also called "leakage" and "passage") with increasing operating time, as available sorption sites are taken (Olsen and Paulson, 2008). NSF/ANSI requires that filters pass twice their rated capacity of contaminated water through the filter, unless there is a performance indication device (PID) that warns the user when capacity has been reached. When a PID is present, the test is conducted to 120 % of the system's rated treatment capacity based on volume. Samples of the challenge water and the treated water are collected at six points throughout the test. On the other hand, some companies producing POU/POE systems use other tests not performed under NSF/ANSI certification protocols, which are usually less stringent: the capacity of the POU/POE filters is not systematically tested. If not properly tested over time, initial results may overlook that the removal performance may decrease with increasing cumulative volumes of water filtered. This may explain the difference in performance compared to some real occurrence studies.

In general, occurrence studies in real conditions should be taken into consideration for a proper evaluation on the efficacy of POU/POE systems. Indeed, while the removal above the maximum capacity has been studied in some lab studies, on the other hand it is conducted in a short time frame and the aging of filters is rarely taken into consideration. These tests are mostly conducted under laboratory-controlled conditions, including constant flow rate (except for Patterson et al., 2019) and water quality (except for Teymoorian et al., 2024), which are not representative of variable household conditions and irregular use patterns. For instance, organic material (TOC) in the influent water affects overall performance of both RO membrane and AC technologies as it accumulates on the media surface (Olsen and Paulson, 2008). This is especially problematic for RO membranes (Herkert et al., 2020; Patterson et al., 2019), which are expected to typically last for at least one year in a residential setting (Olsen and Paulson, 2008). However, such long-term testing in laboratory conditions is hardly feasible for POU filtration systems with a very large theoretical maximum capacity in terms of filtered volume. Some extreme examples have a filter maximum capacity of thousands of litres. This would require testing the system for several years of expected lifetime if only filtering several litres a day.

3.3. Peer-reviewed occurrence studies

All the occurrence studies that included measurement of PFAS in tap water samples are listed in Table 1. In the first seven studies reported in the Table, water samples were taken at the tap; this means, in the presence of a POU/POE treatment, that only post-treatment water was considered. In the other published works, water samples both pre-treatment and post-treatment were analysed, actually evaluating the performance.

The first occurrence study appeared in 2015 and dealt with the analysis of eleven PFAS in samples collected from vending machines, water dispensers and household water filters in Bangkok (Thailand). The results (detection of 4 PFAS) were compared with the values in water from the "water supply pumping station" and direct faucets, where higher concentrations were found (Tabtong et al., 2015).

Guardian et al. also analysed 5 drinking waters from vending machines in Bangkok, as well as bottled waters and source waters close to the city for 33 PFAS. The results for the vending machines were not separately discussed in the article, but a table in the supporting material showed generally lower concentrations for the water samples from vending machines compared to those in the source waters around Bangkok, with the exception of perfluoroundecanoic acid detected in 2 vending machine water samples and in none of the source waters (Guardian et al., 2020).

In the study from Scher et al., samples of outdoor tap water (from municipal systems and homes on private wells), garden soil, and garden produce were collected at homes within a groundwater area contaminated from PFAS landfills in Minnesota and analysed for seven PFAS (Scher et al., 2018). Among the samples, some derived from POU systems treatment. At least one perfluorinated compound (PFBA) was detected in all water samples from unfiltered exterior taps ($n = 17$). None of the seven PFAS were detected from homes on private wells with GAC-filtered exterior taps ($n = 3$).

Ao et al. measured six PFAS compounds (PFOA, PFOS, PFNA, PFBS, PFHpA, and PFHxS) in nine paired tap and filtered water samples served by three different water sources in Shanghai, China (Ao et al., 2019). Concentrations were lower in filtered samples.

Bradley et al. analysed tap waters from 45 homes in the US, including 1 after POU RO, and observed the absence of PFAS in only the POU-RO treated waters. According to the supporting material, 6 of the tap water samples had been collected after passing through POU carbon filters, but these did not seem to have any effect, and the results were not commented by the authors (Bradley et al., 2020).

Likewise, Bradley et al. analysed tap waters from 34 private-well and 22 public-supply sites, including 4 treated by POU adsorptive filters, and 1 by a POE water-softener system. The detailed analytical results can be found in the supporting material, but the authors did not comment on them (Bradley et al., 2022).

In a 2021 survey in Spain, 35 PFAS were targeted from 105 different households without POU/POE treatment ($n = 81$), after AC filters ($n = 14$), and after RO ($n = 10$) in domestic waters in Spain (Cserbik et al., 2024). Four of the five PFAS detected above quantification limits in unfiltered tap water at other households were also detected in AC filtered water, albeit less frequently. No PFAS were detected in RO-treated samples.

Although interesting, these six studies did not focus on the use of POU/POE filters, but just monitored in a rather "basic" way the concentration of the contaminants in treated or non-treated water, also coming from different inputs. No removal efficiency could be calculated and little or no information was given on the type of filters, their age, nor on sampling or quality control in the field. Such basic information is absolutely necessary to interpret the results, limiting the usefulness of these works for further discussion and comparisons.

In the following four studies, water samples were collected pre- and post-POU/POE treatment, thus allowing calculations of removal efficiencies. Herkert et al. were the first to examine the PFAS-removal efficiencies of POU/POE filters in a residential setting (Herkert et al., 2020). Filtered ($n = 89$) and unfiltered ($n = 87$) tap water samples in 73 residences in North Carolina were analysed for 11 PFAS (GenX, PFBS, PFBA, PFHxS, PFOS, perfluoropentanoic acid (PFPA), PFHxA, PFHpA, PFOA, PFNA, and PFDA). RO filters and dual-stage AC filters were found to consistently remove most measured compounds (except PFNA and GenX) at an average of ≥90 % efficiency. GAC filters had more variable performance and were far less effective in removing short-chain PFAS compounds, with an average removal efficiency of just 41 % for those chemicals. AC POE systems resulted in increased levels of PFAS in half of the tests ($n = 4$), which is consistent with some studies regarding PFAS-removal technologies in centralized treatments (Cantoni et al., 2021). Indeed, in some cases, species which are initially adsorbed when more sites are available, but which exhibit low affinity for the sorbent, are later "eluted" causing an apparent increase in concentration.

In another survey in 2020, 35 PFAS were targeted before and after AC filters ($n = 6$) and RO ($n = 5$) in domestic waters in Spain (Cserbik et al., 2023). PFAS were efficiently removed by RO, while AC filters were less efficient, showing once again higher concentrations after filtration

in some samples. The authors hypothesized that domestic AC filters in real-life working conditions did not efficiently adsorb PFAS when highly loaded and clogged, and thus had the potential to release PFAS to the filtered water. Still, no information was collected on filter age to support this hypothesis.

Mulhern et al. sampled 18 different waters from private wells in North Carolina that used commercially available (POU) under-sink AC block filters (strongly packed fine particles of activated carbon), which are certified under NSF P473 protocol for PFOA and PFOS. Samples were taken at approximately monthly intervals for 8 months, two more than the manufacturer's expected lifetime based on time of use. The filters were successful in removing 17 PFAS detected in the filter influent, including short-chain perfluoroalkyl ether acids (PFEAs), over the whole study period (Mulhern et al., 2021). While the volume-based capacity of the tested AC block filters had not been reached after the last sampling, the recommended lifetime based on time of use was deemed to provide a significant safety factor.

He et al. analysed water samples pre- and post-POU treatment using new and unused filters in 12 homes. Commercially available three-stage water purifiers containing AC and RO were evaluated and approximately 60 exhausted filters of different type were also collected to be either extracted and analysed or characterized. Profiles of PFAS in the exhausted POU filters and water samples were built and allowed to assess removal efficiencies and formulate adsorption mechanisms (He et al., 2022). This is so far the only study collecting both filters and water samples for the analysis of PFAS. Overall, these studies in real settings showed that RO POU/POE is effective, while AC POU/POE treatment is less effective and may even lead to increase in the levels of certain PFAS in the treated waters over time.

In contrast to the widely reported water volume (in terms of percentage rated capacity) used in laboratory studies, there was little information on the filtered water volume for filters installed and sampled in regular homes. In most of the occurrence studies, attempts could only be made to relate removal performance to estimates of filter age (i.e., time since installation) when reported. While occurrence studies may have been conducted with older filters more representative of typical residential use compared to laboratory studies, the age of the filters was not reported in some of the studies (Cserbik et al., 2023). To date only Mulhern et al. performed an 8-month longitudinal study covering the manufacturer's expected lifetime based on time of use (recommended filter change every 6 months).

Previous lab and occurrence studies rarely distinguished the removal of PFAS by filters with different functions. Indeed, often removal efficiency were given by analysing waters pre and post-POU/POE treatment. This is not a problem when single-stage filters were used such as in the study by (Mulhern et al., 2021). However, such a method gives little information on the effect of each phase when filters are composed of multiple phases. In fact, POU/POE systems typically consist of a different combination of various filters including filtration, adsorption, IEX and membrane systems. Notably, POU/POE membrane systems often include AC or carbon block prefilters prior to the RO membranes. Previous studies that assessed the removal of PFAS by RO membrane systems rarely differentiate adsorption on carbon block and AC prefilters from RO membrane rejection. This combination of multiple technologies makes it difficult to attribute PFAS removal to specific filters in the POU/POE system. In addition, those multiple filters have different lifespans. Thus, the influence of potentially poor maintenance by the user in real settings on POU performance is even more difficult to assess without proper information.

4. Main current analytical issues, and perspectives in POU/POE studies

4.1. Low quantification limits required for monitoring

A substantial issue in the evaluation of PFAS removal by POU/POE is related to the rather low concentration levels which need to be determined, only attainable with the most recent analytical methods and instrumentations. Concentration limits are generally in the 1–50 ng L^{-1} range, thus, even in the studies with more targeted analytes, few were detected in the influent water. In the studies with the greatest number of PFAS studied (>30 PFAS), the percentage of detected PFAS with respect to the total number considered ranges from 0 % to 64 % (Bradley et al., 2022, 2020; Cserbik et al., 2024, 2023; He et al., 2022; Mulhern et al., 2021; Natarajan et al., 2021; Teymoorian et al., 2024), which could either indicate the absence of the contaminants or the low performance of the analytical method.

According to a recent review, sensitivity for PFOA and PFOS in drinking water analysis has increased by an order of magnitude in the past decade, with detection limits reaching sub-ng L^{-1} values (Teymoorian et al., 2023). Still, not all laboratories are able to measure at such low levels and/or perform appropriate method validation. During the UCMR 3, six PFAS were monitored nationwide between 2013 and 2015 using analytical methods developed by EPA (EPA 537 Rev. 1.1) (EPA Office of Water, 2024a). At the time, the quantitation limits that were considered achievable, with 95 % confidence, by at least 75 % of laboratories nationwide were between 10 and 90 ng L^{-1}. In the latest UCMR (2023–2025 survey), the limits reached by at least 75 % of the laboratories for the same six PFAS were between 3 and 4 ng L^{-1} (EPA Office of Water, 2024b). However, health advisory levels are often even lower. As detection limit requirements go lower with more stringent regulations, it becomes more challenging to develop and validate efficient analytical methods (Teymoorian et al., 2023). Moreover, this challenge is compounded by the ubiquitous presence of PFAS, leading to potential sources of contamination in the laboratory itself and thus affecting the measurement of very low trace levels in samples (Teymoorian et al., 2023). The extreme limits of detection reached in some regulations require not only to use the most cutting-edge instruments, but also efficient sample preparation. Even though the more common and used method, namely SPE, allows preconcentration in a laboratory of several litres of water, this may not be enough to reach the required LODs, also due to the simultaneous pre-concentration of interferents. Research should really be pushed forward to find alternative strategies to cope with these issues, maybe testing more specific materials and novel configurations.

4.2. Limitations in targeted and untargeted mass spectrometric analysis

The other major analytical weakness of the reviewed works (both lab and occurrence studies), with the exception of the most recent studies (2021–2024) is the very low number of PFAS considered (Table 1). In fact, when the scope is limited to verify the performance of POU/POE in removing PFAS, tap water samples are analysed by LC-MS/MS with a typical targeted analysis approach. The focus is generally on the most common contaminants and emerging PFAS are neglected, also due to the difficulty in finding available commercial standards (Ng et al., 2021). By doing so, the evaluation is limited to few compounds, while the huge number of existing PFAS is virtually ignored. The evaluation of the real sample contamination remains thus incomplete. An alternative could be the use of non-target approaches, based on high resolution mass spectrometry (HRMS), which allows the detection, discrimination and tentative identification of a large number of compounds. These strategies, first introduced in metabolomics studies (Menger et al., 2020), have been increasingly applied to different aquatic environmental samples. Nevertheless, drinking water applications have been somewhat neglected due to the expected low concentrations of emerging contaminants (Kutil et al., 2024; Manz et al., 2023).

Among these, two peer-reviewed studies were applied to POU/POE treatment studies, of which one generic and one specifically focused on PFAS. Newton et al. were the first to perform suspect and non-targeted analyses of POU filter extracts from nine North Carolina homes in the USA. Filters were collected when they showed maximum capacity,

which was determined by a change in the indicator colour (after an average time of 68 days). A suspect screening analysis (SSA) consisted of matching the mass spectra of unknowns to a list from EPA's DSSTox database. At the end of the workflow, 15 candidate compounds were ultimately confirmed with the help of available standards and 8 out of the 15 compounds were PFAS (Newton et al., 2018). Non-targeted analysis (NTA) was also performed by McCord et al. on water samples taken pre-, mid-, and post-POE treatment at six private drinking water wells in southwestern New Jersey in 2019. Results showed >90 % PFAS removal, using a semi-quantitation strategy based on the addition of an internal standard (carbon-13 labelled PFNA) (McCord et al., 2020). No information on the number of samples, nor on the two-stage POE systems was reported, except that five were GAC-based POE treatment and one was an anion exchange based POE treatment (McCord et al., 2020).

Even if NTA by LC-HRMS can be a powerful tool, accurate quantitative analysis still represents a challenge in the field. Moreover, the presence of interferents may lead to weaker capacity of detecting contaminants in specific matrices. For example, breakthrough of organic matter from an aged POU/POE filter may lead to higher matrix effects, hindering a reliable evaluation of the removal of non-target compounds. Even if the issue would also be present in the case of target analysis, a check on samples spiked with known concentrations of analytical standards would immediately highlight the problem. On the other hand, in non-target LC-MS approaches, the use of specific standards is not mandatory, making it difficult to estimate if differences among samples arise from varied concentrations or varied matrix effect. For these reasons, along with the very time-consuming and complex characteristics of NTA due to the sheer volume of data generated, this approach on its own may not be the most appropriate to routinely investigate the efficiency/performance of POU/POE systems by analysing pre-treated and treated water samples. Wide-scope targeted approaches could represent a good choice to reliably evaluate POU/POE efficiency for a large number and type of compounds, but with the limitation of high costs related to the purchase of numerous standards and the lack of many reference standards among the huge PFAS family.

4.3. Other potential complementary strategies for water analysis: Non-targeted and surrogate analyses

Some alternative non-target methods may be considered to help the overall assessment of PFAS presence prior and post POU treatments. For example, the Total Organic Fluorine (TOF) methods include the extraction of organic fluorine by SPE or AC columns, a combustion step and the following quantitation of the fluoride ion by ion chromatography (Han et al., 2021). They present the advantages of determining the whole organic fluorine fraction, if compared with LC-MS approaches. Still, the removal of inorganic fluorine must be carefully checked, as well as the recovery of several representative PFAS by the chosen extraction technique (Forster et al., 2023). Another option is represented by the Total Oxidable Precursor (TOP) assay, which involves an oxidative reaction to convert precursors into PFCAs, an SPE procedure and a following LC-MS targeted analysis of well-known short- and long-chain PFCAs (Houtz and Sedlak, 2012). This method smartly overcomes the problem of standard availability but presents the drawbacks of allowing the analysis of solely oxidable compounds, as well as the possibility of non-complete oxidation due to the sample complexity (Ateia et al., 2023). However, these strategies, if correctly implemented, would be useful to evaluate drinking water PFAS contamination before and after POU/POE treatments, upon careful evaluation of the analytical performances.

A few previous studies on full-scale or pilot-scale drinking water treatment (Cantoni et al., 2021; Sgroi et al., 2018) and one laboratory study on POU/POE treatment (Aung et al., 2022) were successful in using UV absorbance at 254 nm and fluorescence to predict removal of PFAS. However, these parameters were mainly reported to be surrogates of the removal of a small number of individual PFAS or the sum of these individual PFAS, and never total PFAS. Still, if good agreement is reached and the breakthrough of the less retained short-chain PFAS can be predicted, easier and cheaper monitoring could be conducted to better understand when to change the filters to avoid doing so too late (unsafe) or too early (uneconomical).

4.4. Extracting POU filters: A revolutionary sampling approach

Ingeniously, POU filters could also be employed as a sampling pre-concentration technique. Indeed, the use of POU filters has recently been advocated and implemented as a sampling medium for microbial monitoring (Bai et al., 2024) and to assess for lead exposure (Pan and Giammar, 2022). This sampling method has the potential to be extended to other inorganic and organic contaminants in tap water, such as PFAS (Johnson et al., 2022). Some of the manufacturers of POU filtration devices claim a removal efficiency of near 100 % for several PFAS and filter lifetimes of dozens of litres to several thousands of litres. If good removal performance is confirmed experimentally, the filters would work as a kind of “in situ” SPE sorbent, integrating the pollution from tap water filtered daily over several weeks or months, resulting in an extremely highly pre-concentrated (dozens of litres to thousands of litres) composite sample. This would be very helpful by sufficiently pre-concentrating PFAS for NTA analysis. In targeted analysis, with knowledge on both the amount of PFAS accumulated on the filter and the volume of water that has passed through the filter, the average concentration in the water during the period can be calculated. This will allow to vastly improve detection limits and consequently permit long-term monitoring while reducing cost and experimental effort by drastically reducing the number of samples to analyse, much like the use of a passive sampler.

So far, the filter extracts were analysed in only 2 studies that included PFAS (He et al., 2022; Newton et al., 2018). However these analyses were not aimed at calculating average concentrations over the filter use period but served different purposes. Newton et al. exploited exhausted POU filter to perform SSA and NTA by Liquid Chromatography-HRMS for ECs (Newton et al., 2018). Thus, by the nature of the analysis, quantification was not possible. In addition, in both studies, sample preparation was not optimized, and mass balance of the retained compounds not assessed. To quantify cumulative PFAS exposure from tap water using POU filters, two requirements must be met. First, the POU filters must exhaustively remove nearly all of the studied PFAS from tap water. Second, it must be possible to extract all of the PFAS that had accumulated on the POU filters. However, there are many gaps in our knowledge concerning uptake of PFAS on POU filters and the processes involved in accumulation and desorption.

4.5. Which POU/POE filter to use?

As previously discussed, only 4 studies actually assessed the removal efficiencies at the POU/POE in real residential settings. Still, by reviewing these studies and with the help of the other occurrence and laboratory studies, some key findings and recommendations can be drawn on the use of AC and RO in POU/POE devices. In general, RO systems seem to more consistently remove PFAS compared to AC systems. Dual stage filters (AC + RO) are also an option: indeed, many RO systems also have the benefit of an additional AC filtration stage included in the system. For private wells, POE RO may be a treatment solution, as the whole house requires water treatment. However, RO suffers from high capital cost, water filtration inefficiency and pipe corrosion. Thus, drinking water that has already been treated by a centralized drinking water treatment plant, should only use POU RO. For AC systems, results vary greatly from very good removal efficiencies to poor removal efficiencies for short-chain PFAS in some cases. A more frequent replacement of the filters may improve the performance but increase the maintenance cost. Indeed, along with the efficiency in PFAS removal, other aspects may be taken into consideration when

recommending the best option, such as costs and waste disposal. Still, these evaluations are currently challenging, due to the lack of literature information.

Many existing POU/POE solutions for PFAS possess very different purchasing and maintenance costs, depending on the brand, type of device and purchase location. To the best of our knowledge, there is no information available in the literature on a rigorous comparative treatment cost comparison between AC, IEX and RO for POU/POE treatment for PFAS in real-life settings. In addition, with rapidly evolving requirements on removal performance, new regulations and costs are expected for waste disposal.

Another consideration is that the management of POU/POE generated waste will depend on local regulations. However, in the US most spent filters end in landfills and RO rejected waters in municipal wastewaters, or in private septic tanks (Alfredo et al., 2023; Patterson et al., 2019). To avoid releasing PFAS back into the environment, future regulations on disposal may be more severe. In that aspect, AC filters are better at concentrating the PFAS and easier to collect and further treat than RO waste waters. Some companies already propose to collect the spent filters.

4.6. POU evaluation: A need for the scientific community

Valuable information on POU/POE performance in removing PFAS may be really helpful in more complex studies regarding for example the presence of fluorinated compounds in biological samples. Since as early as 2006, several researches have measured blood serum levels of PFAS in cohorts and attempted to correlate them with drinking practices – including the use of POU/POE treatment or not – but no water samples were collected nor analysed (Cuffney et al., 2023; DeLuca et al., 2023; Emmett et al., 2006; Siebenaler et al., 2017; Tian et al., 2018; Zhou et al., 2019). When statistically significant correlations could be observed among PFAS levels in blood and use of POU/POE, concentrations were often higher for those that had no POU/POE treatment at home. For example, lower levels of the long-chain PFOA was associated with the use of POU/POE treatment (Cuffney et al., 2023; Emmett et al., 2006; Siebenaler et al., 2017). In a large US study, drinking municipal water treated by home filters were associated with lower PFAS concentrations in human serum for all seven PFAS tested, compared to drinking municipal water without additional filtration (Cuffney et al., 2023). In few cases, a higher concentration of some specific PFAS was measured in the blood serum of those who used POU/POE treatment (DeLuca et al., 2023; Siebenaler et al., 2017). Nonetheless, it must be pointed out that PFAS intake may derive from different sources, thus direct conclusions on the issue are difficult to draw. In addition, there is no information on the type of POU/POE treatment devices in place; some do not treat PFAS, and others may even release PFAS in the treated waters. Thus, better knowledge on the POU/POE systems in place, and on actual PFAS concentrations in POU/POE treated waters may help in relating serum levels of PFAS to drinking water habits.

5. Conclusions

From the few studies found and reviewed, it emerges that a deep investigation on the removal of PFAS by POU/POE treatments is still lacking. Nonetheless, their evaluation is becoming necessary, due to the spreading of such devices and ubiquitous PFAS contamination.

Some POU/POE treatment systems have been tested and certified to remove certain PFAS. In addition, some companies on their website indicate the removal percentages for many PFAS by their POU/POE systems according to outside laboratories. However, only few occurrence studies (real life scenarios) have been carried out at the tap so far, and only five of these assessed removal efficiencies.

Not all water filters are effective at removing PFAS, especially short-chain PFAS, and some AC systems may even increase the PFAS concentration in purified waters if not properly maintained. Current studies appear to support RO as the best treatment for PFAS separation, but since the disposal of the contaminated rejected water is problematic, AC is better for concentration and management of the PFAS waste. However, much more data is needed to fully assess the performance of POU/POE treatment towards PFAS removal, especially in real-life settings. For a reliable recommendation on the most suitable filter type, removal efficiency is a crucial aspect but costs, future regulations and final fate of the removed PFAS should not be neglected.

The different tests performed when not following NSF/ANSI guidelines make it difficult to compare the results between filters. Often, it is not known how long a given treatment system maintains the reported level of performance (water volume-wise or time-wise). Only occurrence studies can allow the verification of performance over time and continuous use and are currently needed to try to draw a picture of the exposure to these contaminants by treated-water intake.

CRediT authorship contribution statement

Henry MacKeown: Writing – original draft, Investigation, Conceptualization. **Emanuele Magi:** Writing – review & editing. **Marina Di Carro:** Writing – review & editing. **Barbara Benedetti:** Writing – original draft, Investigation.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.176764.

Data availability

Data will be made available on request.

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