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Poly- and perfluoroalkyl substances in water and wastewater: A comprehensive review from sources to remediation

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Abstract

Per- and polyfluoroalkyl substances (PFAS) are pollutants have attracted major concern due to their high persistence and bioaccumulation. They are causing increasingly serious epidemiological problems in many communities globally due to consuming PFAS-contaminated water sources. Necessarily, the behavior of PFAS in water and wastewater needs to be understood better. This study attempts to comprehensively review, analyze and discuss PFAS based on the following key aspects: (i) sources, (ii) occurrence in water and wastewater, (iii) transformation, fate and migration, and (iv) remediation technologies. Studies indicated that modern water and wastewater treatment plants cannot deal completely with PFAS and in some cases, the removal efficiency is minus -3500-fold. The main reasons are the high hydrophobicity of PFAS and presence of PFAS precursors. Precursors can account for 33–63% of total PFAS concentration in water and wastewater. Detection and identification of precursors are challenging due to the requirement of advanced analytical instrument and standard chemicals. Several technologies have been developed for PFAS remediation involving two main mechanisms: separation-concentration and destruction. The most widespread in-use technology is adsorption because it is reasonably affordable. Anion exchange resin and synthesized materials are the most effective sorbents having a sorption capacity of 100–2000 mg PFAS/g sorbent, effective within a few hours. The destruction technology such as plasma can also be a promising one for degrading PFAS to below health-based standard in 1 min. However, plasma is costly and not yet ready for full scale application.

Abbreviations: AFFF, aqueous fire-fighting foam; AOP, advanced oxidation process; diPAP, DiPhosphate esters; DWTPs, drinking water treatment plants; EPA, Environmental Protection Agency; F-53B, potassium salt of 6:2 chlorinated polyfluorinated ether sulfonate; FASE, perfluoroalkane sulfonamidoethanols; FOSA, perfluorooctane sulfonamide; FOSAA, perfluorooctane sulfonamidoacetic acid; FOSE, perfluorooctane sulfonamidoethanol; FP, fluoropolymers; FTAC, fluorotelomer acrylate; FTCA, fluorotelomer carboxylic acids; FTOH, fluorotelomer alcohols; FTSA, fluorotelomer sulfonic acids; PAP, fluorotelomer phosphates; PASF, perfluoroalkane sulfonyl fluoride; PFAA, perfluoroalkyl acids; PFAS, per- and polyfluoroalkyl substances; PFBA, perfluorobutanoic acid; PFBS, perfluorobutane sulfonic acid; PFCA, perfluoroalkyl carboxylic acids; PFDA, perfluorodecanoic acid; PFDoDA, perfluorododecanoic acid; PFDS, perfluorodecane sulfonic acid; PFHpA, perfluoroheptanoic acid; PFHpS, perfluoroheptane sulfonic acid; PFHxA, perfluorohexanoic acid; PFHxDA, perfluorohexadecanoate; PFHxS, perfluorohexane sulfonic acid; PFNA, perfluorononanoic acid; PFNS, perfluorononane sulfonic acid; PFOA, perfluorooctanoic acid; PFOS, perfluorooctane sulfonic acid; PFFPA, perfluoroalkyl phosphonic acids; PFPE, polymeric perfluoropolyethers; PFPeA, perfluoropentanoic acid; PFPeS, perfluoropentane sulfonic acid; PFPiA, perfluoroalkyl phosphinic acids; PFSA, perfluoroalkylated sulfonate; PFTeDA, perfluorotetradecanoate; PFTTrDA, perfluorotridecanoic acid; PFUnDA, perfluoroundecanoic acid; POSF, perfluorooctane sulfonyl fluoride; WWTPs, wastewater treatment plants

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1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are fluorine-based compounds that were first manufactured in the 1940s. PFAS molecules possess fluorine atoms which replace the hydrogen atoms partly or entirely, and include a functional group (e.g., sulfonate and carboxyl) [1]. The fluorine atoms impact on the properties of the alkyl chain which make it more compact, dense and hydrophobic. The bonding of fluorine and carbon atoms possesses low polarity and high strong bond energy, up to 536 kJ/mol [1]. PFAS have been used globally for water- and oil-repelling purposes, such as coating on kitchen cooking ware, food, packaging, wax, paint and aqueous fire-fighting foams (AFFFs). PFAS are applied widely thanks to their specific chemical structures. The increasing use of PFAS for both domestic and industrial applications has resulted in serious environmental issues. PFAS are now detected in various environmental matrices, specifically water and wastewater. For instance, in Australia the average concentration of 21 PFAS in 19 wastewater treatment plants (WWTPs) reaches 110 ng/L [2]. The recommended concentration according to the National Environmental Management Plan (Australia) is only 0.23 ng/L, which is virtually 500-fold lower [2]. It suggests the current water and WWTPs are unable to deal with this problem. In the last ten years, countries such as the US, Australia, EU and recently China, have established guidelines and conducted institutional and academic strategies to combat PFAS.

In the effort to remove PFAS from so many industrial processes, several technologies have been developed based on two mechanisms: separation-concentration and destruction. For example, adsorbents from traditional forms (e.g., activated carbon, anion exchange resin) to the advanced ones (e.g., imprinted polymer, colloidal and amide-functionalized sorbent) have been tested to separate PFAS from water and wastewater [3–7]. For the destruction mechanism, advanced oxidation process (AOP), thermal and plasma technologies have been done [8–10]. However, what they achieve is still modest. The main problem stems from the refractory and hydrophobicity of C–F bonding, in which PFAS can resist most technologies. Another reason is the complicated transformation and detection of PFAS. The precursors of PFAS are widespread in the environment and they can transform into terminal PFAS products. Dealing with PFAS precursors is another problem. After performing treatment, the long chain PFAS can break up into short chain PFAS which are much more difficult to remove compared to the former. The final objective of PFAS removal is to defluorinate the C–F bonding; however, this is an expensive process requiring a sophisticated system, such as AOP, thermal and plasma [8,9]. None of those technologies are yet ready for full-scale application in the market place.

PFAS are disposed of in water and wastewater sources at worryingly high levels, up to 120 µg/L [11]. They enter the food chain and drinking water, subsequently exposing people and creating an epidemiological problem [12]. PFAS are biologically resistant compounds and the C–F chain can resist biological degradation. Therefore, in the human body, PFAS accumulate in organs and blood serum after several years of consuming PFAS-contact food, water and using PFAS-based products. Recent reports have linked PFAS with low infant birth weight, thyroid hormone disruption, low activity sperm, diabetes and cancer, especially in the blood serum of firefighters who are exposed to PFAS frequently [12].

Some review studies on PFAS have been published but there is still much more to do. For instance, Mahinroosta and Senevirathna [13] reviewed treatment technologies for remediating PFAS in soil. Likewise, Gagliano et al. [14] examined the possibility of PFAS adsorption by carbonaceous materials. Li et al. [15] and Ateia et al. [16] looked at the literature on short chain PFAS. More broadly, Lu et al. [17] investigated the treatment train technologies for ex-situ conditions. These above-mentioned studies focused on particular aspects of PFAS-relevant issue, such as remediation technologies (e.g., sorption), typical sorbent types (e.g., carbonaceous, amide-functionalized sorbent), PFAS groups (e.g., short and ultrashort chain) and environmental matrix (e.g., soil).

However, PFAS in water and wastewater bodies have not been comprehensively studied. Several reasons indicate the importance of reviewing PFAS in water and wastewater. The water and wastewater matrix closely relates to other matrices. Applying soil washing for PFAS remediation generates huge amounts of wastewater. Industry and domestic activities also discharge PFAS wastewater that require proper solutions. In practice, the long chain and short chain PFAS co-occur and compete with each other. In terms of water chemistry, PFAS present in real water and wastewater in a more sophisticated way compared to laboratory conditions. Laboratory experiments use only distilled water but real water and wastewater contain several interferences (e.g., organic matter). This makes the remediation process difficult and challenging. To address those issues, the knowledge of PFAS in water and wastewater matrix must be reviewed in detail.

Hence, this study attempts a comprehensive review, data analysis and discussion of PFAS based on the following key aspects: (i) sources, (ii) occurrence in water and wastewater, (iii) transformation, fate and migration, and (iv) remediation technologies. This review is expected to serve as a reference for future analyses on the subject. The data of PFAS are collected from studies published since 2008, with a focus on water and wastewater sources in countries such as the US, Australia, EU, Canada, China, Japan, and South Korea.

2. Classification, properties and sources of PFAS in water and wastewater

2.1. Classification and properties of PFAS

PFAS is an abbreviation for a large fluorine-based group that includes a set of subgroups. Currently, 4300 PFAS are registered in the Chemical Abstracts Service. Thus, it is necessary to understand the classification of PFAS for further usage. PFAS are categorized as polymer and non-polymer substances. In the non-polymer group, per-fluoroalkyl acids (PFAA) is the most commonly known subgroup. PFAA subgroup includes perfluoroalkyl carboxylic acid (PFCA) and per-fluoroalkylated sulfonate acid (PFSA) (Fig. 1). PFOA is a typical PFAS compound of the subgroup PFCA. Similarly, PFOS is the well-known one in the subgroup PFSA. The polymer PFAS are formed by monomer and divided into three subgroups, these being fluoropolymers, polymeric perfluoropolyethers, and side-chain fluorinated polymers. They can break up into terminal PFAA products so they are also considered PFAA's precursors. The precursors are perfluoroalkyl which are linked with replaceable groups such as silane, alcohol, amide, epoxide, cyanide, sulfide and metal.

* The selected PFAS compounds in PFSA and PFCA subgroups are the subject of this review. The detailed classification of PFAS is documented in the study by Buck et al. [18].

PFAA are a major class of PFAS which appear in most research studies and government health-based guidelines. They have added to the momentum of monitoring and remediating PFAS in water and wastewater. This review study focuses on selected compounds in PFAA class and their precursors for the above reasons so as it is important to understand thoroughly the naming convention of this class. When naming PFAA, there are two regular ways to do so and these are based on: (i) functional groups; and (ii) chain lengths counted on the number of C atoms in the chain.

To date four functional groups have been identified in the PFAA class. Counting the number of C atoms in the chain is one convention for naming PFAS, typically PFBA (C4) and PFODA (C18). PFAS are subdivided into short chain and long chain. The number of C atoms which determines the short or long chain differs amongst the functional groups. The relevant EU authorities define that the short chain of PFCA consists of less than seven perfluorinated carbons. The short chain of PFSA holds fewer than six perfluorinated carbons. The long chain PFCA contains seven or more perfluorinated carbons and the corresponding PFSA possesses six or more perfluorinated carbons. In contrast, the US

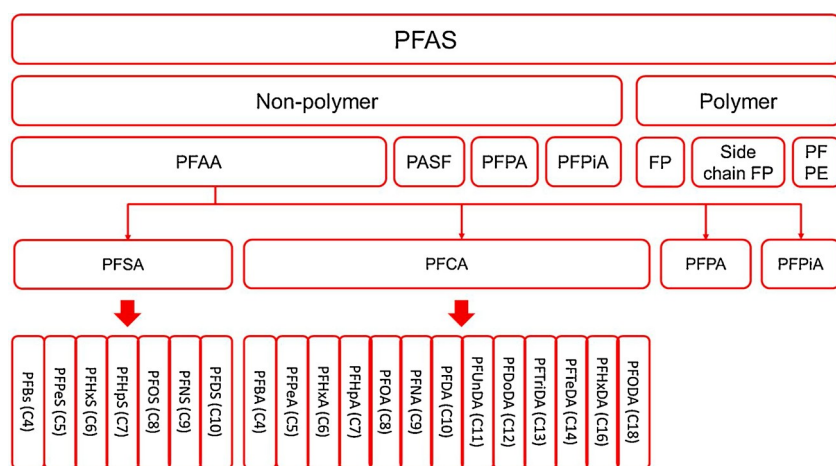


Fig. 1. Simplified PFAS classification.

Environmental Protection Agency (EPA) regulates that short chain PFCA contains less than eight carbon atoms and PFSA has less than six carbon atoms.

Most long chain PFAS contain more than eight carbons in the chain. Their molecules' weight is higher than 400 g/mol and this results in high hydrophobicity of long chain PFAS. For example, the solubility varies from -1.93 to -6.68 mol/L and $\log K_{ow}$ ranges from 3.16 to 5.3. The short chain PFAS have been substituting for long chain PFAS in recent years for industrial applications. The short chain PFAS are more hydrophilic and soluble than the long chain variety. The solubility of short chain PFAS ranges from -2.9 to 0.94 mol/L and this is three times higher than the values of long chain PFAS. Similarly, the $\log K_{ow}$ of short chain PFAS is less, ranging from 0.5 to 4.6. In other words, the bioaccumulation potential of short chain PFAS in humans and biota is less than long chain PFAS. This explains why short chain PFAS have replaced long chain PFAS to prevent epidemiological problems. The long chain PFAS of the sulfonate group have the greatest toxic effect on biota, while short chain PFAS accumulate 5–600-fold less [19].

Apart from the short chain PFAS, replacement PFAS are another option to phase out long chain PFAS. The popular replacement PFAS in market include Gen X, Adona and F53-B. They are the replacement version of PFOA serving for fluoropolymer. Generally, the replacement PFAS possess 7–8 C in the chain and differ from traditional PFAS. Their structure comprises potassium salt and oxygen which link up in the middle of the chain. The oxygen bonding is easily encountered and the replacement PFAS is broken up into smaller fractions. However, it has been reported that the replacement PFAS is just as toxic as the ordinary PFAS due to the comparable bioaccumulation rate [20].

2.2. Sources of PFAS

Given that PFAS have been used in many scenarios in recent years, PFAS appear in various sources in different inventories. According to US EPA, the most common ones can be found in: (i) food packaging materials, (ii) commercial household products, (iii) workplaces, (iv) drinking water, and (v) living organisms. This section focuses on the first three environments as they contribute significantly to PFAS in water and wastewater. The occurrence of PFAS in drinking water is described in Section 3.4.

Food-packaging materials are the largest source of PFAS. They are manufactured by coating PFAS on the surface of materials to increase the water-repellent, oil-proofing characteristics of the material and slow down the food decaying process. PFAS-packaging material is mostly used in the fast-food industry. Paper tableware, popcorn bags, cupcake cups, instant noodle cups, beverage containers and baking paper are the main products (Table 1). After use, the materials are disposed of and

dumped in landfill. Landfill leachate enters the groundwater or surface water and introduces PFAS to the water sources [21,22]. This problem has been reported in several advanced economies like the US, EU and Australia, but not much is known in developing countries, including China, Thailand, India and Egypt.

The composition and concentration of PFAS in those materials differ significantly from country to country. For example, FtOH concentration in packaging bags in the US and China is significantly different [23]. The reason for this is because the telomerization method has served as the main strategy for synthesis since 2003. An intensive screening of PFAS in food-packaging materials has been conducted in 17 countries [24]. It emerges that Greece had the highest PFAS level in its products, following by India and China. This discrepancy is attributed to the current legislation and guidelines in place, which allow PFAS to be coated on packaging materials at different levels. It has potentially impacted on the PFAS concentration in water and wastewater sources in several nations.

The next emerging PFAS source constitutes commercial household products, for example stain- and water-proofing, non-stick, cleaning and AFFF materials. AFFF is the most relevant to water and wastewater. It contains substantial amounts of PFAS compounds using PFCA, PFSA, switterionic, fluorotelomer and other novel unidentified classes. It depends on the manufacturer's adjustment of AFFF formulation. For example, the old generation AFFF in Norway contains substantial PFOS, but the newer one must have less than 0.001 %, as legislated by the EU [25]. The new AFFF has mainly 6:2 FTSA (40–800 mg/L) and small doses of other PFAS. In contrast, the AFFF brands in France (e.g., 3 M, Ansul, Chemguard, Buckeye) contain very large PFCA concentrations, ranging from 22.5 to 3188 mg/L [26]. Those PFAS sources penetrate through the soil layer in the grounds of military bases and fire stations, and enter groundwater much deeper below. The AFFF which attaches on fire-fighters' clothes also contributes to PFAS in wastewater after the washing water is discharged [27,28].

Other household equipment also contains PFAS, such as water proofing agents, impregnation agents, cleansers, polishes, lubricants, paints, ski-waxes, inks and tanning substances [25,29,30]. The frequently-detected PFAS substances in household equipment belong to PFCA and PFSA groups. Unlike AFFF, the FtOH group is not generally found in household items or equipment. The only item which contains FtOH is the impregnation agent. It possesses long chain FtOH ranging from 0.9 to 1.8 g/L. The concentrations of other PFAS in household equipment are extremely high, at similar level as in AFFF. For example, the concentrations of PFOS in the foam suppression agent can reach 22.7 g/L, which is 36-fold higher than in AFFF (0.63 g/L) [29]. After using the cans, boxes and bottles which contain left-over PFAS are disposed in landfill. In the same way, PFAS are introduced to water

Table 1
Sources of PFAS.

Category	Items	Studied countries	Important subclass	Selective PFAS compound	Concentration	Sampling time	Reference
Food packaging	Paper table ware, popcorn bag, cupcake cup, paper cup, paper box, paper bag	China and US	FtOH	6:2 FTOH 8:2 FTOH 10:2 FTOH 12:2 FTOH 14:2 FTOH 16:2 FTOH 18:2 FTOH	(< 0.60–1110 ng/g) (< 0.40–8490 ng/g) (< 0.02–9350 ng/g) < 0.02–8450 ng/g < 0.02–1640 ng/g <0.02–372 ng/g <0.02–130 ng/g	2013 - 2015	[23]
	French fries cardboard box Sandwich wrapping papers, cardboard pasta containers, unprocessed baking cups	Egypt	PAP, PFSA and PFCA	6:2 and 8:2 monoPAP PFOA PFOS PFHxS and PFDS	Above the method detection limit 2.40 ng/g 0.29 ng/g Below detection limit	2013	[32]
	Grease proof paper, Oven safe paper, take away cup (board), thick paper bag, baking paper, burger box and French fries box	Sweden	PAPs	Mono- and di-PAPs (6:2 - 14:2)	Detected but no data of concentration	2012	[33]
	Instant noodle cup, microwave-popcorn bag, beverage cup, ice cream cup, fast-food container, dessert container, baking paper	Thailand	PAP	PFOS PFOA	92.48 ng/dm ² 16.91 ng/dm ²	2011 - 2012	[34]
	Food contact paper, paper board, paper cup, beverage container	US	PFCA, PFSA, FTSA, PAP, PF ether	PFBA, PFPeA, PFHxA, PFOA, PFBS, PFHxS, 4:2 FTSA, 6:2 FTSA, 10:2 FTSA	16-800 nmol/cm ²	2014 - 2015	[35]
Commercial household products	Waterproofing agent, AFFF	Norway, Sweden	PFCA, PFSA, FtOH	PFBS PFOS PFOA PFBA 6:2 FtOH 8:2 FtOH 10:2 FtOH	38.65-253700 µg/L 568000 µg/L 26-208 µg/L 81 - 27647 µg/L 535-13250 µg/L 26500 - 54780 µg/L 5800 - 120721 µg/L	2009	[25]
	AFFF (3 M brand)	US	PFCA, PFSA, Switterionic, Fluorotelomer and novel classes	N-SP-FASA, N-TAmP-FASA, N-FASAP, CMAmB-FA	Qualitative, not quantitative	2011 - 2015	[28]
	Impregnation agents, cleansers, polishes, lubricants, foam-suppressing agents, paints, ski wax, inks and tanning substances	Switzerland	PFCA PFSA	PFPeA (C5) PFHxA (C6) PFHpA (C7) PFOA (C8) PFNA (C9) PFDA (C10) PFBS (C4) PFHxA (C6) PFOS (C8) ipPFNS (C9) PFDS (C10) N-MeFOSE (C11)	5.3 mg/L 0.6-6.2 mg/L 5.6 mg/L 15.2 mg/L 0.2 mg/L 0.1 mg/L 26.8 mg/L 89.5 mg/L 634.9 mg/L 5.1 mg/L 3.2 mg/L 0.3 mg/L	2012 - 2013	[29]
	Household equipment (textile, floor covering, plastic); building material (wood, insulation material, façade material, air-conditioning)	Czech Republic	FASE PFSA, PFCA		0.033-77.6 ng/g	2016	[30]
	AFFF concentrate	France	FTCA, FTSA	6:2 FTSA 6:2 FTBA	5-78.5 mg/L 5-3188 mg/L	2015 - 2016	[11]

Workplace	Chrome plating wastewater	China	Replacement PFAS	6:2 FtSaAM F-53B	50-125 mg/L 65.7 µg/L	2018	[31]
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Table 2

Occurrence (ng/L) and removal efficiency (%) of selected PFAS in WWTPs in various countries.

Categories	Selected compounds	Sampling sites	Influent concentration (ng/L)	Effluent concentration (ng/L)	Removal efficiency (%)	Sampling time	References
PFCA	PFBA (C4)	Australia, France, China, Sweden, US, Spain, South Korea	0.45 - 199605	0.33 - 11942	-277 - 94	2010 - 2018	[2,11,36,37,38,39,40,41,42,43,44,45,46,47,48]
	PFPeA (C5)	Australia, France, US, China, Japan, Thailand, Sweden, Uganda, Spain	1.49 - 228944	1.35 - 15299	-745 - 93	2010 - 2018	[2,11,36,37,38,39,40,42,44,45,46,47,49,50]
	PFHxA (C6)	Australia, France, US, Japan, Thailand, Kenya, Sweden, Uganda, China, Spain, South Korea	0.3 - 689260	0.2 - 47689	-1536 - 93	2010 - 2018	[2,11,36,37,38,39,40,41,42,43,44,45,46,47,48,49,50,51,52]
	PFHpA (C7)	Australia, France, US, China, Kenya, Sweden, Spain, South Korea	0.51 - 143792	0.2 - 11658	-1467 - 92	2010 - 2018	[2,11,36,37,38,39,40,41,42,43,44,45,46,47,48,49,50,51,52]
	PFOA (C8)	Australia, France, US, China, Japan, Thailand, Kenya, Sweden, Denmark, Uganda, Spain, South Korea	0.9 - 120592	1.4 - 12759	-460 - 89	2010 - 2018	[2,11,36,37,38,39,40,41,42,43,44,45,46,47,48,49,50,51,52]
	PFNA (C9)	Australia, France, US, China, Japan, Thailand, Kenya, Sweden, Denmark, Uganda, Spain, South Korea	0.78 - 3500	0.37 - < 4000	-200 - 37	2010 - 2018	[2,11,36,37,38,39,40,41,42,43,44,45,46,47,48,49,50,51,52]
	PFDA (C10)	Australia, France, US, China, Japan, Thailand, Kenya, Sweden, Denmark, Uganda, Spain, South Korea	0.3 - 4292	0.16 - < 4000	-3624 - 100	2010 - 2018	[2,11,36,37,38,39,40,41,42,43,44,45,46,47,48,49,50,51,52]
	PFUnDA (C11)	France, Japan, Thailand, Kenya, US, China, South Korea	0.3 - 946	0.4 - < 4000	-323 - 82	2013 - 2016	[11,40,42,44,48,49,50,51]
	PFDODA (C12)	Australia, Japan, Thailand, Kenya, US, France, China, South Korea	0.03 - 2684	0.11 - < 4000	-267 - 73	2013 - 2015	[11,48,49,50,51,52]
	PFTriDA (C13)	France, China, Spain, South Korea	0.59 - < 5000	0.02 - < 10000	-100 - 99.8	2013 - 2015	[11,47,48,50]
	PFTeDA (C14)	France, China, Spain, South Korea	0.02 - < 5000	0.02 - < 10000	-100 - 100	2013 - 2015	[11,47,48,50]
	PFHxDA (C16)	China, Spain, South Korea	0.04 - 0.31	0.04	0	2013 - 2017	[47,50]
	PFODA (C18)	Spain	300	190	37	2014 - 2017	[47]
	PFSA	PFBS (C4)	Australia, France, US, China, Sweden, Uganda, China, Spain, South Korea	0.21 - 204440	0.11 - < 4,000	-1829 - 98	2012 - 2018
PFPeS (C5)		Australia, Sweden	0.3 - 1.76	0.3 - 2.82	-67 - 13	2014 - 2017	[2,43]
PFHxS (C6)		Australia, France, US, China, Japan, Thailand, Kenya, Denmark, US, Spain, South Korea	0.3 - 559973	0.37 - 34518	-80 - 94	2012 - 2017	[2,11,37,39,40,42,43,45,46,47,48,49,50,51,52,53]
PFHpS (C7)		Australia, France, China, US, Sweden, Spain	0.19 - 37221	0.17 - < 4000	-33 - 89	2010 - 2017	[2,11,37,39,43,47]
PFOS (C8)		Australia, France, US, China, Japan, Thailand, Kenya, Sweden, Denmark, Uganda, Spain, South Korea	0.4 - 1907840	1.4 - 29876	-225 - 98	2010 - 2018	[2,11,36,37,38,39,40,41,42,43,44,45,46,47,48,49,50,51,52]
PFNS (C9)		Spain	3.28 - 5.62	0.04	99	2010 - 2011	[47]
PFDS (C10)		Australia, France, US, Sweden, China, Spain, South Korea	0.1 - 524	0.34 - < 4000	-663 - 100	2014 - 2017	[2,11,38,41,46,47,48,50]
FTSA	Replacement PFAS						
	Precursor						
FTCA							

4:2 FTSA (C6)	-	2013 - 2016	[11,36,44,46]				
	6:2 FTSA (C8)	Australia, France, US, Sweden	7.29 - 337260	6.13 - 83200	-100 - 65	2012 - 2017	[2,11,36,41,43,46]
France, US	8:2 FTSA (C10)	Australia, France, Sweden	0.1 - 24200	0.1 - 5081	-10 - 79	2012 - 2017	[2,11,36,43,44,46]
	6:2 FTCA (C8)	France	2.8 - 5100	< 1,000	80	2013 - 2016	[11,44]
0.8 - 400	8:2 FTCA (C10)	France	870	< 1,000	-15	2015 - 2016	[11]
	10:2 FTCA (C12)	France	< 1,000	< 1,000	~100	2015 - 2016	[11]
2 - 400							
	F-53B (C8)	China	65700	-	-	2018	[31]
	FOSA (C8)	France, US, Kenya, Sweden, Denmark, China, Spain	0.9 - 49937	0.056 - < 10,000	-1485 - 100	2010 - 2016	[11,36,38,46,47,50,51,53]

(continued on next page)

Table 2 (continued)

Categories	Selected compounds	Sampling sites	Influent concentration (ng/L)	Effluent concentration (ng/L)	Removal efficiency (%)	Sampling time	References
FtOH	FOSAA (C8)	France, US, China, South Korea	1.37 - < 5,000	0.14 - < 10,000	-100 - 90	2014 - 2016	[1,3,6,46,48,50]
	6:2 FTOH (C8)	France	110	55.35	10.88	2013	[44]
	8:2 FTOH (C10)	France	4,265	340	12.53	2013	[44]
	10:2 FTOH (C12)	France	1,820	344	14.07	2013	[44]
	12:2 FTOH (C14)	France	1.12	0.26	0.12	2013	[44]

The removal efficiency is calculated by the following equation (when not present in literature): Removal efficiency (%) = $(C_{in} - C_{out})/C_{in}$.

sources through leaching from landfill.

The last source of PFAS is the workplace encompassing production facilities and various industries. Typical activities are chrome plating, electronics manufacturing or oil recovery. The chrome plating wastewater in Jiangsu province (China) has the replacement PFAS compound (F-53B) at 65.7 µg/L [31]. Similarly, F-53B is detected at 43–78 µg/L in the chrome plating’s industry’s wastewater effluent in Wenzhou City, China [20]. Details concerning this PFAS source in wastewater are presented in Section 3.1.

3. Occurrence of PFASs in water and wastewater environment

In this section, we summarize the occurrence of PFAS in various water and wastewater environments. We categorize those environments into four kinds: WWTPs, surface water, groundwater and drinking water. The data are analyzed according to the concentration range of selected PFAS compounds in various countries. Data are also evaluated statistically to compare the occurrence of each PFAS compound and total PFAS concentration amongst the reported countries.

3.1. Occurrence in wastewater treatment plants

The information concerning selected PFAS in WWTPs is summarized in Table 2. The data include influent, effluent concentration and removal efficiency of selected PFAS such as PFCA, PFSA, FTSA, FTA, replacement PFAS, precursor and FtOH, which appear in many WWTPs. Based on the published information, we analyze the data statistically to elucidate and discuss two important issues: firstly, the huge difference between minimum and maximum PFAS concentration; and secondly, the current trend of using PFAS.

The difference between minimum and maximum values of every PFAS in either influent or effluent is very large (Table 2). For instance, the values of PFPeA differ 2.3 million-fold. Similarly, the concentrations of minimum and maximum PFBS in the PFSA group are one million times different. This discrepancy can be explained by the origin of upstream wastewater sources whether from domestic wastewater, or PFAS non-intensive or intensive industrial wastewater. That is, the minimal values of PFAS concentration are attributed to domestic wastewater and the maximal values links to industrial wastewater (Table 3). For example, the concentration of PFAS in effluent of WWTPs, downstream from the textiles industry, varies from 232 to 1115 ng/L [37,53]. This value is much higher than that of domestic wastewater which is below 20 ng/L. The WWTPs, downstream of the AFFF industry and firefighting training grounds, receive exceptionally high PFAS concentrations compared to other industries, up to 700,000 ng/L [11]. In Table 2, the maximum PFAS concentration originates from the wastewater of firefighting training grounds [11]. In those areas, the total concentrations of PFAS were 5-fold higher than the corresponding values in domestic wastewater [37]. This contrast indicates that the marginal concentration of PFAS in WWTPs mainly depends on the wastewater type of the upstream sources.

Apart from the AFFF industry, other PFAS-discharged sources include paint, commercial surfactant concentrate, water proofing agents and chrome plating. The data reported in Table 1 confirm the scenario showing extremely high PFAS concentrations in those products. Thus, we can extrapolate that the corresponding wastewater from those industries carry similarly high PFAS loads. Another notable PFAS source found in WWTPs is hospital wastewater [51]. PFAS in hospital wastewater originates from medical devices like radio-opaque, in vitro diagnostic and color filters [51]. Based on the literature, we can classify the range of total PFAS concentrations in WWTPs as shown in Table 3 below.

The last concern of PFAS in WWTPs is which compounds appear in the highest concentration and detection frequency. In general, the concentration of short chain PFAS is at least 50-fold larger than the long chain (Table 2). The concentration of short chain PFAS is always higher

Table 3
Range of PFAS concentration in wastewater.

Sources	PFAS concentration range (ng/L)	Typical products	References
Domestic	> 100	Food packaging, dust, household equipment	[23,25]
PFAS non-intensive industry	100-1000	Chrome plating, hospital	[31,51]
PFAS intensive industry	> 1000	Water proofing agents, AFFF	[14,20,27]

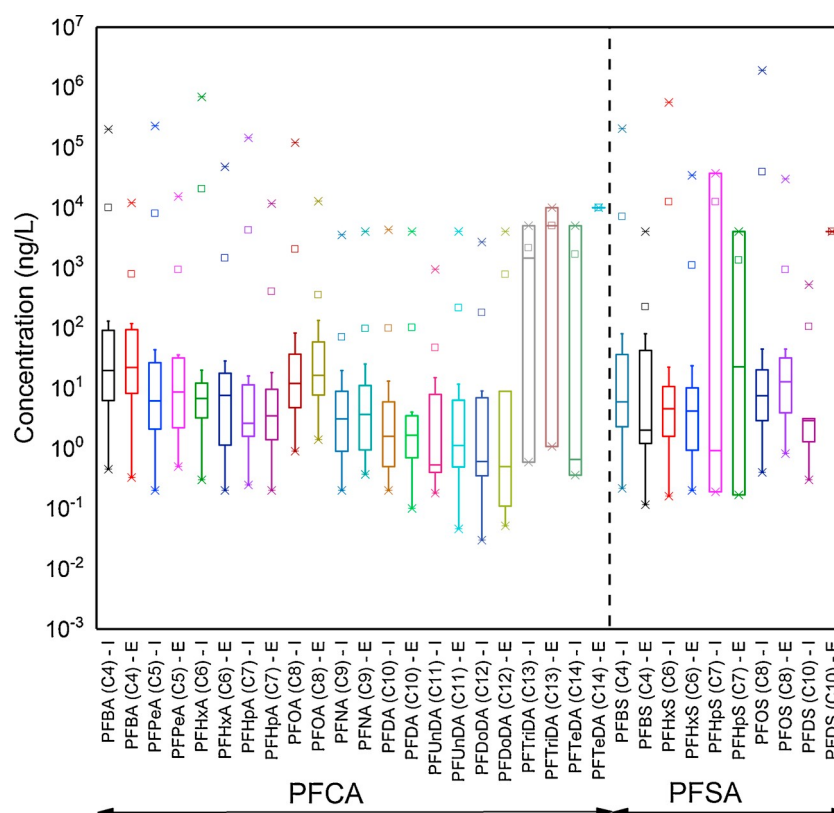


Fig. 2. Influent and effluent concentrations (ng/L) of selected PFAS compounds in PFCA and PFSA groups in WWTPs. Performed data is from Table 2. The formulation of labels in vertical axis comprises PFAS name – C chain length – sampling position. “I” stands for influent. “E” stands for effluent.

than 10⁶ ng/L whereas long chain PFAS is below 5000 ng/L (Fig. 2). This reflects the current trend of using short chain PFAS for manufacturing PFAS products. However, the situation complies with the policies and regulations of each government. The EU, US and Australia have been replacing long chain PFAS since 2000 by using PFBS, PFBA and 6:2 FTSA. In Australia, PFBA is the most widely used and its highest concentration is 370 ng/L. The detection frequency of 6:2 FTSA also reaches 99 % in WWTPs. Australian industries have been using PFBA and 6:2 FTSA as C8 to replace AFFF and metal plating [2]. However, some countries such as Kenya and Uganda still favor the old ones. The most commonly detected PFAS in Kenya are PFOA and PFOS but short chain PFAS do not present in most samples [51].

3.2. Occurrence in surface water

After treatment in WWTPs, PFAS are released to surface water and distributed into water sources. The data of PFAS distribution in surface water are summarized in Table 4. Most of the reported PFAS compounds belong to PFCA, PFSA, FTSA, FTCA and precursors. Unlike the case of WWTPs, PFAS appear in surface water at a lower concentration range (Fig. 3a). This is because the PFAS mass has been diluted in surface water and can be affected by rainfall. In Vietnam, a surface water collected in the two biggest rivers in the north and south has a rather low total PFAS concentration, below 5 ng/L [54]. Similarly, total PFAS concentration in Marseille Bay (France) ranges from 0.11 to 9 ng/L

[55]. To date, PFAS in surface water have been reported in countries like Canada, France, Sweden, China, Vietnam, Australia and Uganda (Fig. 3b). The total PFAS concentration in Canada and China is 1 log magnitude higher than other countries. Especially, the value in France exceeds others three log magnitude. The surface water source in the case of France receives waste stream from AFFF-related industry [11,55]. That explain why the total PFAS concentration of France is significantly high.

We can also see that concentration of short chain is higher than that of long chain. Specifically, the mean concentration of the short chain PFCA group varies from 1 to 10 ng/L while the long chain ranges from 0.01 to 1 ng/L. In Cambridge Bay (Canada), PFOA, PFNA, PFDA, PFOS and PFBS have been detected [56]. The short chain PFBS is the dominant compound (14.7–85.1 ng/L), followed by PFOA (9.92–80.4 ng/L) and then PFOS (4.20–70.6 ng/L). In Lake Victoria (Uganda), PFBS is the dominant PFAS (91 %) at a concentration of 2.3 ng/L [57]. In Lake Victoria (Uganda), PFBS is the dominant PFAS (91 %) at a concentration of 2.3 ng/L [42]. PFOA and PFOS still present at a significant level, emphasizing the fact that they are still preferred in various industries. However, we did not exclude the possibility of long chain PFAS being redistributed from sludge and sediment of lakes and rivers. More data for the PFAS profile in the sediment and sludge must be explained. The redistribution of long chain PFAS after snow and ice melted is a typical example [56].

The distribution of the PFAS profile in surface water is impacted by

Table 4
Occurrence (ng/L) of PFAS in surface water.

Categories	Selected compounds	Sampling sites							
PFCA		Canada	France	China	Australia	Sweden	Vietnam	Finland	Uganda
	PFBA (C4)		19601	0.32 – 1.1		1.55 – 1.97		5.3	< Lod
	PFPeA (C5)		18130	0.36 – 0.55		0.974 – 1.34		4.2	< Lod
	PFHxA (C6)		193 – 70560	0.27 – 0.64	4.4 – 13	1.2 – 1.29	0.04 – 0.52	5.7	0.3 – 1.5
	PFHpA (C7)		16912	0.19 – 0.75	1.2 – 6.2	0.635 – 0.656	0.05 – 1.22	2.7	< Lod – 1.5
	PFOA (C8)	9.92–80.4	0.18 – 23604	0.48 – 5	6.1 – 22	1.22 – 1.41	0.25 – 2.28	5.4	0.6 – 2.4
	PFNA (C9)	2.33 – 11.7	< 400	0.04 – 0.37	1.1 – 3.3	0.249 – 0.286	0.06 – 0.3	23	0.1 – 0.4
	PFDA (C10)	0.44 – 4.67	569	0.02 – 0.32	1.2 – 1.4	0.092 – 0.108	0.01 – 0.13	0.52	< Lod
	PFUnDA (C11)		< 400	0.02 – 0.07		0.03 – 0.042	0.01 – 0.06	1.9	< Lod
	PFDoDA (C12)		< 400	0.02 – 0.03		< Lod – 0.032	0.03 – 0.04	< Lod	
	PFTriDA (C13)		< 1,000	0.02 – 0.14			0.03 – 0.05	<Lod	
	PFTeDA (C14)		< 1,000	n.d.			< 0.06	< Lod	
	PFHxDA (C16)							0.43	
	PFSA	PFBS (C4)	14.7–85.1	94608	0.08 – 1.3	n.d – 6.4	1.33 – 1.55	0.12 – 0.31	1.5
PFPeS (C5)									< Lod - 1.2
PFHxS (C6)			0.15 – 219312	0.12 – 0.29	1.5 – 36	1.62 – 2	0.01 – 0.47	6.4	
PFHpS (C7)			16490					0.4	
PFOS (C8)		4.20–70.6	0.25 – 892320	0.54 – 4.99	2.2 – 38	2.95 – 3.89	0.01 – 0.41	26	< Lod – 1.6
PFDS (C10)			< 400			< Lod	< 0.11	< Lod	
FTSA	4:2 FTSA (C6)		< 400					< Lod	
	6:2 FTSA (C8)		119808					2.7	
	8:2 FTSA (C10)		8968					0.13	
FTCA	6:2 FTCA (C8)		1040						
	8:2 FTCA (C10)		< 1,000						
	10:2 FTCA (C12)		< 1,000						
Precursor	FOSA (C8)		2336			0.025 – 0.048		0.7	
	FOSAA (C8)		< 1,000						< Lod
Total PFAS concentration (ng/L)		54.7 – 248	12451 – 7654819	0.74 – 110	33.6 – 137.7	0.51 – 14.29	0.23 – 14.6	0.13 – 26	1.1 – 14
Sampling time		2005 – 2018	2015 – 2018	2014 – 2015	2017 – 2018	2014 – 2015	2010	2014	2015
References		[56,58]	[11,55]	[37,59]	[60]	[38]	[54]	[57]	[42]

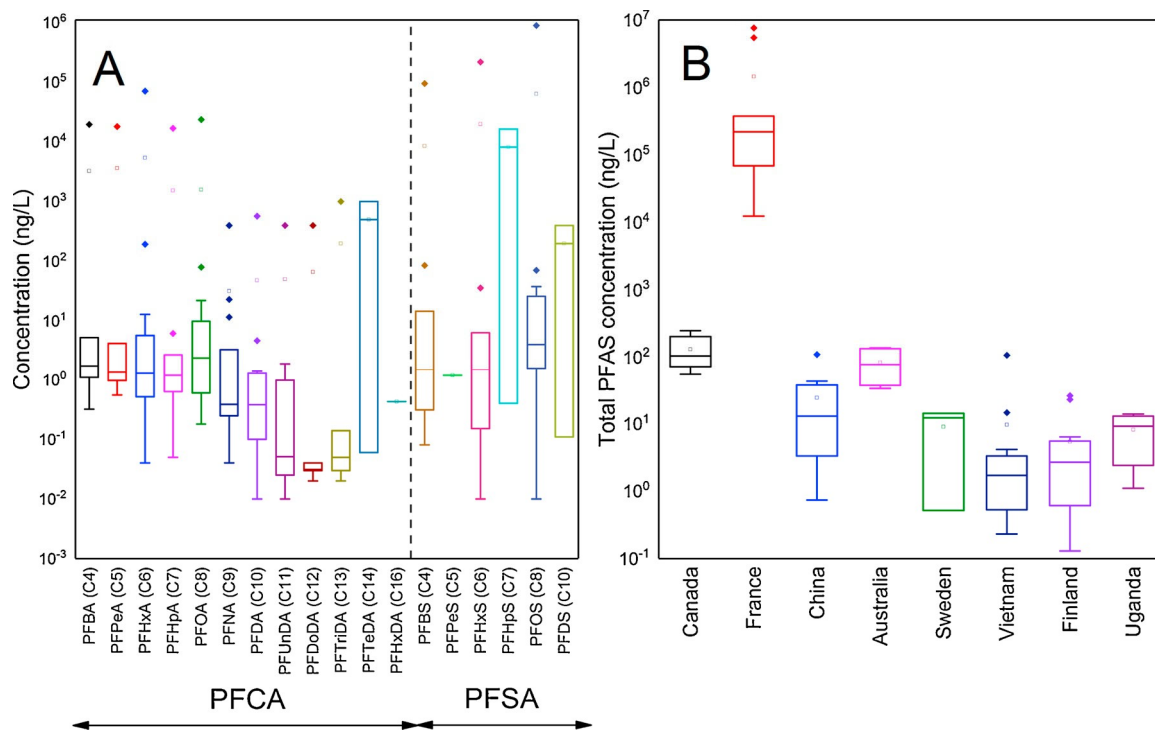


Fig. 3. Concentration of selected PFAS compounds in PFCA and PFSA groups in surface water (ng/L) (A) and in various countries (B). Performed data is from [Table 4](#). The formulation of labels in the vertical axis illustrated in [Fig. 3a](#) comprises PFAS name – C chain length.

the following critical factors: types of surface water (e.g., river or sea), temporal and spatial ones. Referring to the types of surface water, river water sources are more influential than sea water [55]. In France, the Rhône River contains a total PFAS concentration of 200 ng/L whereas Marseille Bay records only 9 ng/L. The underlying reason is that PFAS mass in the river has been diluted before it enters the sea. We also speculate that PFAS have been partly remediated in the river and adsorbed in the sediments prior to reaching the sea. Regarding the effect of temporal variance, the dry season receives the highest PFAS concentration, followed by medium and wet seasons. In China, the total PFAS concentrations in the dry, medium and wet seasons range from 4.8 to 37.6 ng/L, 12.2 to 110 ng/L and 3.3 to 43.0 ng/L, respectively [59]. The high temperature in the dry season speeds up the degradation of precursors and in turn increases the PFAS concentration in the water matrix. During the wet season, rainfall dilutes the PFAS concentration and this may also be affected by the maintenance schedules of local industries in dry and wet seasons, such as boat repairs. Thus, the amount of grease repellent and paint containing PFAS falls during certain times of the year. However, no matter what the season, the short chain PFAS and PFOA are the dominant compounds reported in several cases [59,61,62].

3.3. Occurrence in ground water

Groundwater is the water source most impacted by PFAS. Similar to wastewater and surface water, a variety of PFAS compounds have been detected in groundwater (Table 5). The detected concentration of PFAS in groundwater is apparently higher than in surface water. From Fig. 4a, the mean concentration of PFAS ranges from 1 to 100 ng/L which exceeds 2 log magnitude than in surface water. The concentration of PFAS in groundwater is notably high because it is directly influenced by the upstream sources. We can categorize the upstream

sources of PFAS in groundwater into high strength and low strength ones.

The high strength sources encompass AFFF-related products such as military bases, airports and firefighting training grounds. At those places, AFFF directly seeps into groundwater or adsorbs to the cement surface, being washed away by rain and then entering the groundwater somewhere else. The PFAS concentration attributed to this source is significantly high, at least 1000 ng/L. For instance, the total PFAS concentration at Williamtown Airforce Base (Australia) reaches 200,000 ng/L [63]. The concentration of PFOS is the highest at 100,000 ng/L. In China, PFBS and PFOA concentration of industry-impacted groundwater ranges from 2000 to 20,000 ng/L [64]. Also, the PFBS level gradually increases 24-fold from 872 to 21,000 ng/L from 2009 to 2017. This indicates the replacement of short chain PFAS for industries has been implemented in these countries.

For the low strength source, the concentration of PFAS reportedly stays below 100 ng/L. The most relevant low strength source includes non-industrial zones, river-based, coastal and WWTP-related and landfill areas. In Jiangsu province (China), PFAS concentration in the southern parts (Nantong and Taizhou) exceeds the northern and central region (Nanjing) which houses the industrial zones [65]. Most PFAS concentrations stay below 10 ng/L. Similarly, the reported concentration of PFAS in coastal, WWTP-related and landfill areas does not surpass 100 ng/L [22,66,67]. However, the seasonal factor is also important. In summer, when the groundwater level is low, the concentration of PFAS reaches its maximum and vice versa [22].

Concentrations of short chain PFAS in ground water surpass the long chain one at least by 1 log magnitude (Fig. 4b). In Fuxin (China), PFBS is the dominant compound governing 79–84 % of total PFAS, while PFOA contributes 49–68 % [64]. Likewise, PFBA and PFBS are detected at 98 % and 83 %, respectively, of which 40 % is higher than the long chain PFAS [65]. PFOS (96 %), PFBS (86 %), PFOA (82 %) and

Table 5

Occurrence (ng/L) of PFAS in groundwater.

Categories	Selected compounds	Sampling sites			
PFCA		Australia	Malta	China	France
	PFBA (C4)	0.01 – 11		0.25 76	2.9 – 327
	PFPeA (C5)			0.76 – 24	8.2 – 902
	PFHxA (C6)	3.7 – 50	1.95	4.64 – 614	7.3 – 1340
	PFHpA (C7)	0.8 – 8.4	1.36	< 0.1 – 766	3.3–224
	PFOA (C8)	2.2 – 12	2.68	1.2 – 2510	1.1 – 341
	PFNA (C9)	0.4 – 0.73	0.9	< 0.1 – 1.06	< 4 – < 40
	PFDA (C10)	0.31 – 1.8	n.d	< 0.1 – 0.5	< 4 – < 40
	PFUnDA (C11)	< 0.2		< 0.3 – 28.9	< 4 – < 40
	PFDoDA (C12)			0.36 – 1.22	< 4 – < 40
	PFTriDA (C13)				< 10 – <100
	PFTeDA (C14)			0.55 – 2.28	< 10 – <100
	PFHxDA (C16)			< 0.5	
PFSA	PFBS (C4)	3.19 – 9		21.2	1.3–750
	PFPeS (C5)	1 – 6.3			
	PFHxS (C6)	5.9 – 34	2.22	0.715 – 2.2	4.6–2860
	PFHpS (C7)	4.4 – 4.59			< 4 – 204
	PFOS (C8)	11 – 129.12	2.09	0.403 – 20.2	2.4 – 539
	PFDS (C10)	0.22			2.1 – < 40
FTSA	4:2 FTSA (C6)				< 4 – < 40
	6:2 FTSA (C8)	1.3		0.44 – 2.83	< 4 – 790
	8:2 FTSA (C10)				< 4 – < 40
FTA	6:2 FTCA (C8)				< 10
	8:2 FTCA (C10)				< 10 – <500
	10:2 FTCA (C12)				< 10 – <500
Precursor	FOSA (C8)				< 10 – <500
	FOSAA (C8)				< 10 – < 500
Total PFAS		0.03– 244000	1.2 – 11.2	2.69 – 26700	4 – 8277
Sampling time		2016 – 2019	2019	2009 – 2015	2012 – 2016
References		[22,63,66]	[68]	[64,65]	[67,69]

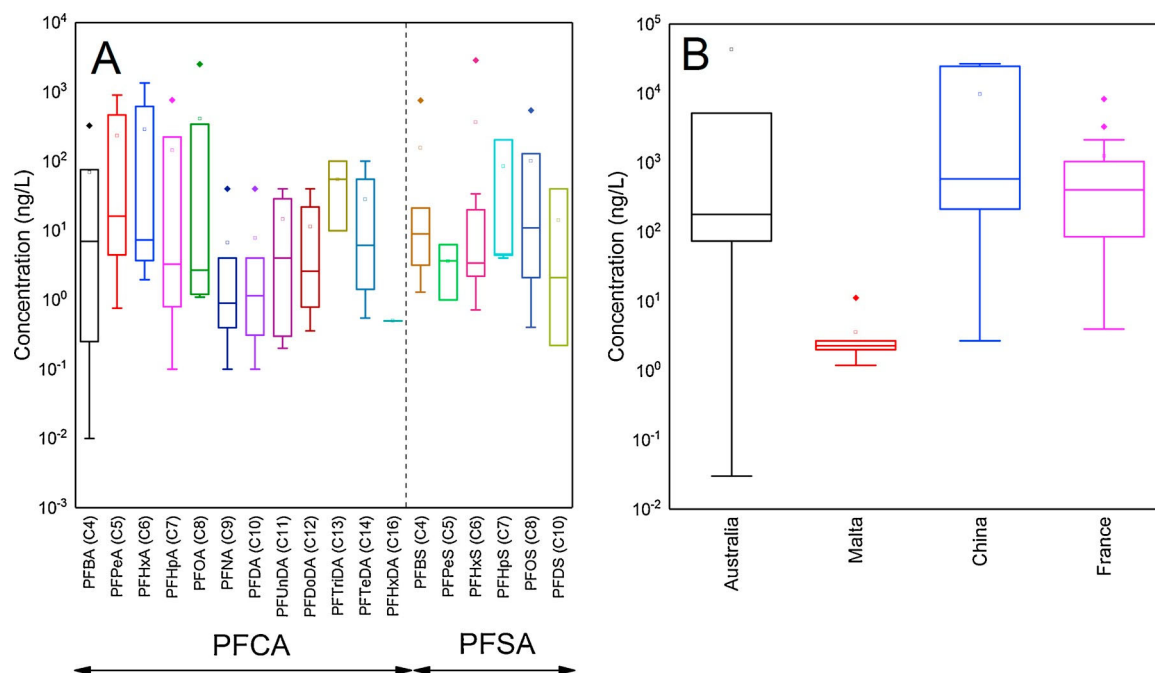


Fig. 4. Concentration of selected PFAS compounds in PFCA and PFSA groups in groundwater (ng/L) (A) and in various countries (B). Performed data is from Table 5. The formulation of labels in the vertical axis illustrated in Fig. 4a comprises PFAS name – C chain length.

PFBA (77 %) are also the most frequently detected compounds in Australia’s groundwater [66]. The most probable reason for this is that the long chain PFAS have been adsorbed by the soil. Only the short chain PFAS having high hydrophilic characteristics can seep through the soil and enter groundwater sources.

3.4. Occurrence in drinking water

Occurrence of PFAS in drinking water is summarized in Table 6. Substantial PFAS groups are detected in drinking water (e.g., PFSA,

PFCA, FTSA and precursor). The mean concentrations of those PFAS range from 0.1 to more than 1 ng/L which is lower than groundwater 2 log magnitude, but very similar to the level of surface water (Fig. 5a). The concentrations of short chain and long chain PFAS consistently correlate. The concentration of short chain PFAS is 1 log magnitude higher than long chain PFAS. Meanwhile, PFOA, PFBA, PFOS and PFBS are the most frequently detected compounds in drinking water. For instance, PFOS contributes 35–40 % of total PFAS in those DWTPs [37]. Also, in DWTPs in Queensland (Australia), PFOA and PFOS are detected at the highest concentrations of 6.7 and 15 ng/L, respectively. The short

Table 6

Occurrence (ng/L) of PFAS in drinking water.

Categories	Selected compounds	Sampling sites					
PFCA		China	Sweden	Vietnam	US	South Korea	Canada
	PFBA (C4)	0.43 – 1.6	1.55 – 1.97		104		1.3 – 3.6
	PFPeA (C5)	0.36 – 0.55	0.974 – 1.34		514	5.51	1.1 – 2.7
	PFHxA (C6)	0.24 – 0.64	1.2 – 1.29	< 0.52	60.8	5.52	0.58 – 4.5
	PFHpA (C7)	0.09 – 0.75	0.635 – 0.656	0.07	177	2.72	1.1 – 3.2
	PFOA (C8)	0.28 – 5	1.22 – 1.41	0.14	108	5.83	3 – 4.9
	PFNA (C9)	0.1 – 0.37	0.249 – 0.286	0.07	38.6	0.873	0.05 – 4.5
	PFDA (C10)	n.d. – 0.14	0.092 – 0.108	< 0.03	24.7	0.441	0.69 – 1
	PFUnDA (C11)		0.03 – 0.042	< 0.06	1.85	0.152	0.39 – 1.6
	PFDoDA (C12)		0.032, < lod	< 0.04	0.09	n.d.	1.1 – 2.9
	PFTriDA (C13)			< 0.05		n.d.	0.17 – 0.94
	PFTeDA (C14)			< 0.06	n.d.	n.d.	0.08 – 0.62
	PFHxDA (C16)				n.d.		n.d.
	PFSA	PFBS (C4)		1.33, 1.55	0.3	11.9	2.603
PFPeS (C5)						15.1	
PFHxS (C6)			2, 1.62	0.02	38.4	0.713	0.67 – 1
PFHpS (C7)						n.d.	
PFOS (C8)			3.89, 2.95	< 0.03	36.9		0.67 – 4.1
PFDS (C10)			< lod	< 0.11	n.d.		1.5
FTSA	6:2 FTSA (C8)						1.3 – 6.3
Precursor	FOSA (C8)		0.025, 0.048				< lod – 1.1
Total PFAS		15.5 – 243	12.2 – 14.2	1.19 – 2.15	1.42 – 1101.73	1.44 – 133	0.59 – 44
Sampling time		2014 – 2015	2013	2013 – 2015	2017	2017	2015

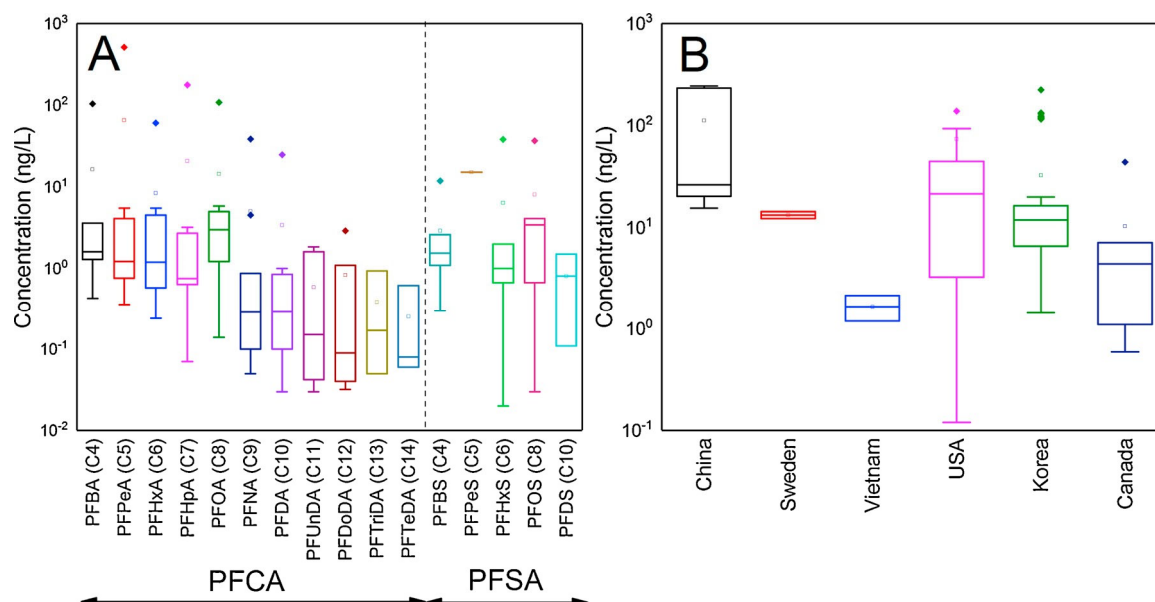


Fig. 5. Concentration of PFAS in drinking water (ng/L) (A) and in selected countries (B). Performed data is from Table 6. The formulation of labels in the vertical axis illustrated in Fig. 5a comprises PFAS name – C chain length.

chain PFBA and PFBS contribute 16–26 % of total PFAS and this discrepancy is the same in other countries like Vietnam, Canada, South Korea and the US [54,70–72]. However, it should be noted that the total PFAS concentration in drinking water in those countries is very different (Fig. 5b). China records the highest total PFAS concentration, up to 100 ng/L while Vietnam has the lowest value of 1 ng/L. Other countries, specifically the US, Canada, Sweden and South Korea possess 1–10 ng/L. This data can partly reflect the current PFAS contamination and management situations in those nations. Nevertheless, we do not exclude the effects of external factors such as temporal and seasonal ones. As previously mentioned in the context of surface water, PFAS concentrations in the dry and wet seasons are 5 to 10-fold different. DWTPs which receive those surface water sources will produce significantly affected drinking water.

Another notable issue is that DWTPs are less effective in PFAS removal [37,72]. The PFAS concentration in tap water is found to be 10–40-fold higher than its original sources in lakes [72]. The reason is due to the precursor in the water sources. From Table 6, the concentration of precursors (e.g., FOSA) ranges from 0.025 to 1.1 ng/L. During the water treatment process, ozonation which occurs in the DWTPs degrades the precursors into terminal products such as PFNS and PFOS [37]. Furthermore, the technologies employed in DWTPs (e.g., sand filtration, flocculation, sedimentation) perform poorly in removing PFAS. For example, the concentration of PFOA and PFOS exposed to granular activated carbon in the effluent is 0.28–0.44 ng/L, whereas it is 4.2–5.2 ng/L using powder activated carbon. Those sorbents can adsorb long chain PFAS but have proven to be useless for short chain PFAS [37].

4. Transformation, fate and migration of PFASs in water and wastewater

4.1. Transformation of PFAS in water and wastewater

The phrase ‘transformation of PFAS precursors’ means they are transformable to PFAS terminal products. Precursors have been detected in most water and wastewater matrices (Table 7) and they influence a huge part of total PFAS mass. In wastewater, precursors dominate 33–63 % of total PFAS concentrations [36]. The most frequently detected precursors include FOSA, FOSAA, FtOH and FTSA, which are able to transform to PFOA and PFOS. The concentration of

precursors in wastewater matrix stays between 10⁻¹ to 10⁴ ng/L, depending on sampling sites. The primary precursor source includes AFFF; thus, water and wastewater collected near AFFF-infected grounds contain more precursors. In surface water, the total concentration of PFOS precursors is 0.025–0.7 ng/L. FOSA, FOSAA, methyl and ethyl derivatives and 6:2/6:2 diPAP (diPhosphate esters) have been detected [73]. In groundwater, concentration of precursors is 100 times larger than in the surface water, being 500 ng/L due to the leachate of AFFF to the groundwater source.

The precursors can be transformed to PFAS through two main routes, these being biotransformation and oxidation. The pathways of selected PFAS precursors transformation are depicted in Fig. 6. For biotransformation, it is driven by microbes and depends on the types of microbes involved [79–81]. For example in aerobic conditions, FtOH is transformed quickly in 48 h [76] but PFOSAmS is degraded slowly, taking 6 months for PFOS when using *Chlorobi* [82]. The driven catalysts are enzymes (e.g., *Cannabis sativa* L. protein) in specific environments [63,77]. For instance, 6:2 FtTAoS can be degraded in sulfate-reducing conditions, to 6:2 FtTP [77]. The transformation of those precursors can increase total PFAS concentration by 38 % [78]. For each PFAS compound, masses of PFHxA, PFOA, PFHxS and PFOS increase by 83 %, 28 %, 37 % and 58 %, respectively [43].

The transformation of precursors via oxidation mechanism happens in similar ways but undertaken by hydroxyl radical groups (%OH). This route is subjected to the AOP and plasma remediation technologies. More details can be found in Sections 5.4 and 5.5. Compared to biotransformation, the oxidation of precursors reacts at a faster rate. More non-toxic terminal products are detected via the oxidation pathway, such as CH₃COOH, HCOOH, F⁻, and SO₄²⁻ [8]. The probably reason is oxidative reagents which encounter C–F chain and defluorinate PFAS thanks to the high energy potential involved. In turn, biotransformation just breaks the functional groups and the C–C chain. There is no intensive defluorination being carried out through the process of biotransformation.

4.2. Fate of PFAS in water and wastewater

Another aspect is the fate of PFAS. As previously mentioned in Section 3, most of the current WWTPs and DWTPs are ineffective in PFAS and precursors remediation. For instance, in DWTPs operating in Australia, the mean concentration of \sum_{21} PFAS shifted from influent to

Table 7

Selected precursors found in DWTPs and WWTPs and their terminal products.

Matrix	Precursor	Major products (% of precursor)	Sampling time	Reference
WWTP	6:2 FTSA	PFPeA (1.5%), PFHxA (1.1%)	2011	[74]
WWTP	6:2 FTSA	6:2 FTOH (6.2%), 5:3 FTCA (17.4%)	2013	[75]
WWTP	6:2, 8:2 FTOH	PFHxA, PFPeA, PFOA, PFHpA	2018	[76]
WWTP	PAP, FTSA	PFHxA (83%), PFOA (28%), PFHxS (37%), PFOS (58%)	2012 – 2015	[43]
WWTP	6:2 FtOH	PFCA (1.7%)	2015	[36]
WWTP	FOSA	PFHxA, PFOA	2013	[38]
WWTP and soil	6:2 FtTAoS	6:2 FtTP	2018	[77]
Surface water	FOSA, FOSAA	PFOS (74%)	2013 – 2014	[73]
Surface water	Unknown	PFCA (38%), PFEA (99%)	2014 – 2018	[78]
Ground water	6:2 FTSA	Short chain PFCA	2012 – 2013	[69]

final effluent and recycled water, at 76, 140 and 120 ng/L, respectively [2]. The poor removal efficiency in those treatment plants is typical of most PFAS groups. In the PFCA group, the PFDA has a removal efficiency of -3624 %, while in the PFSA group, PFBS is removed at -1829 % (Fig. 7). The precursors transform to terminal PFAS and increase PFAS mass in WWTPs. Subsequently, they undermined any efficiency in the removal of PFAS.

Most of those plants are designed to incorporate three treatment stages: primary, secondary and tertiary steps (optional) using a wide range of physical, chemical and biological technologies. Those processes generate sludge, and PFAS are detected in sludge samples, at a frequency beyond 90 % [2]. The mean PFAS concentration in sludge samples stays at 34 ng/g, meaning that part of the PFAS mass has partitioned in the sludge, especially the long chain compound. However, the concentration of PFAS in effluent still higher than in the influent of treatment plants (Table 2). It confirms that the amount of transformed precursors dominates the amount of PFAS adsorbed in sludge. Other processes in treatment plants, such as biological treatment, chemical treatment, simply serve to transform PFAS precursors, rather than degrade PFAS. For instance, FOSA, FOSAA and FtOH break down to PFOS, PFOA and PFHxA through the bioprocess in WWTPs [84,85]. The precursor POSF-based perfluorochemicals and FTOH degrade to PFOA and PFOS [53].

The second reason for the poor removal efficiency is due to the historical use of PFAS. The previously used PFAS adhere to the concrete surfaces of firefighting pads and sewage systems until they leach into a WWTP [11]. Those residual PFAS might transform into the end products and give rise to a large PFAS concentration in the effluent of WWTP. Also, PFOS, PFHxS, and PFBS can remobilize from historical sources (e.g., airports, firetrucks) and end up in the WWTPs [36]. Another possible cause is that PFAS partition into sludge in WWTP, and then are released into the effluent [48]. Technical designs and

operations may also lead to certain outcomes. The effluents from WWTPs and reject water of the reverse osmosis process become mixed. The reject water contains substantial long chain PFAS that would increase PFAS concentration in the final effluent [2].

4.3. Migration of PFAS in water and wastewater

The migration of PFAS in water and wastewater is still not adequately understood due to constraints in sampling and interference of various factors. Two main routes of PFAS migration in water and wastewater comprise: (i) discharged sources to groundwater; and (ii) WWTPs to surface water. The groundwater can bring PFAS to stream, surface water and enter the ocean. The transportation from discharged sources (e.g., airports, firefighting training grounds and landfill) to groundwater is governed by the characteristics of the soil, including organic matter and mineral. It determines how much and what kind of PFAS are retained in the soil layer via the hydrophobic, electrostatic interaction, ion exchange and hydrogen bonding [13]. Not only can sand and soil layers hold 80 % and 32 % PFOS, respectively [86], they also determine the level of PFAS at the downgradient sites. For example, at a distance of 500 m the PFAS concentration is equivalent to 0.001 to 0.1 of the initial concentration [67]. Each PFAS compound is also distributed differently in the soil layers. Short chain PFAS have less retardation than the long chain.

Referring to the second route, PFAS migrate from the WWTPs to surface water via a river and ultimately end up in the ocean. Then PFAS are transported by currents to the polar regions where they are frozen and become part of the icebergs [87]. The behavior of PFAS distribution in this route is determined by water depth, organic matter concentration and microbial activity. At the surface, 80 % of total PFAS is PFCA but at the deeper layers, PFCA and PFSA contribute 55 % and 42 %, respectively [88]. PFAS have been found at a depth of 3500 m in the

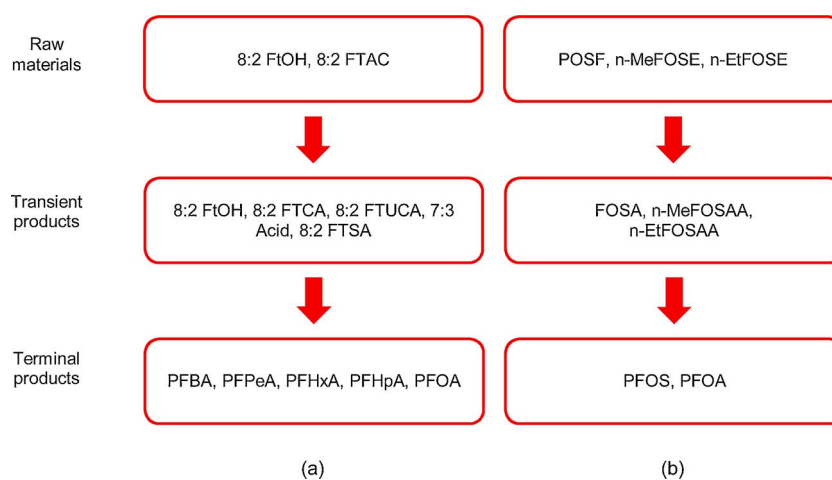


Fig. 6. Transformation pathways of selected PFAS precursors: Fluorotelomer (a) and perfluorooctane sulfonyl (b). Modified from ITRC [83].

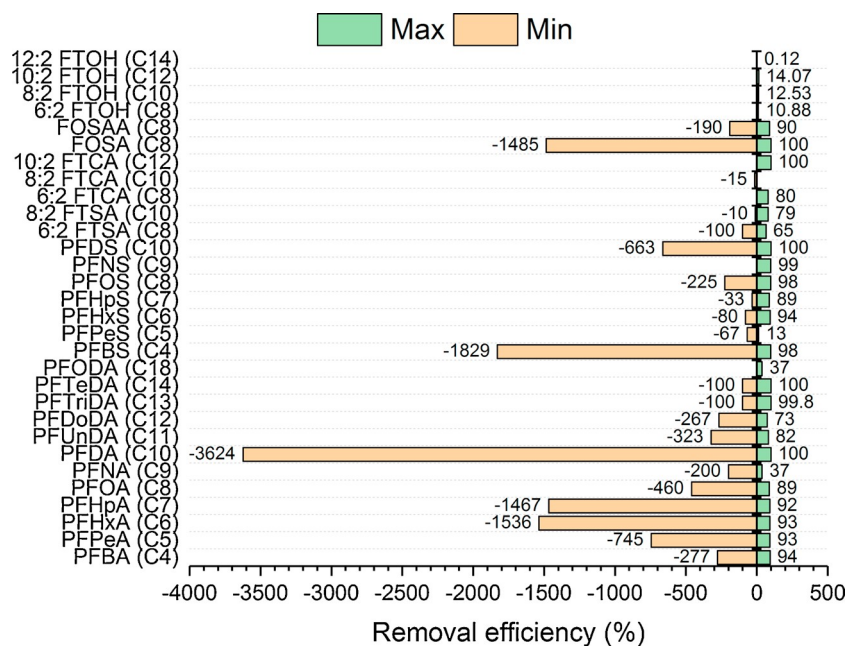


Fig. 7. Removal efficiency (%) of selected PFAS in WWTP. Performed data is from Table 2. The formulation of labels in the horizontal axis comprises PFAS name – C chain length.

Labrador Sea [87]. In general, the PFAS concentration appears to be smaller in the deeper layers. Regarding the effect of organic matter, FTSA and PFSA are retained similarly while the polyfluoroalkyl betaines are absorbed more than PFCA [89]. Also, PFAS are impacted by bacterial activity but more studies are needed on this issue [88].

Transformation, fate and migration of PFAS in water and waste-water remain a huge challenge for management and remediation. The reason stems from the detection and identification of PFAS and their precursors which require advanced analytical instruments such as quadruple time of flight and high-resolution mass spectrometry. As well, standard chemicals for the precursors are still limited in the market place and they are expensive. Most analyses conducted on PFAS precursors are qualitative but more quantitative studies are required. Several PFAS precursors still present and migrate in water and waste-water but have not yet been adequately identified.

5. Treatment process of PFAS remediation in water and wastewater

To date, a variety of technologies has been developed to remediate PFAS in water and wastewater sources. We can classify the technologies into two types: firstly, separation-concentration; and secondly, destruction. The separation and concentration technologies include adsorption, stabilization and solidification and membrane. The destruction ones comprise AOP, thermal, sonochemical, bioprocess and plasma. The primary purpose of the first category technologies is to separate and concentrate PFAS to condensed concentration. Then, the condensed PFAS solute is defluorinated by the second category technologies. Those technologies are sequential aspects of a sophisticated PFAS remediation system.

5.1. Separation and concentration technologies

5.1.1. Adsorption

Adsorption has emerged as a key technology in PFAS remediation. Traditional adsorbents include carbonaceous material and anion exchange resins. Activated carbon remains the most common form of adsorption for PFAS remediation [6]. Nevertheless, the performance of carbonaceous adsorbents and anion exchange resin is compromised by

co-contaminants such as anions and organics. Consequently, advanced technologies or strategies have been developed, such as polymer-based and synthesized materials. Those adsorbents are modified progressively to increase PFAS adsorption capacity based on the affinity and porosity of adsorbent. For example, activated carbon has been fabricated as single walled carbon nanotubes and microporous activated carbon [90]. The adsorption capacity of those materials is different (Table 8). The synthesized adsorbent and anion exchange resin surpasses the carbonaceous and polymer ones (Fig. 8a). The adsorption capacity of carbonaceous material ranges from 10 to 100 mg PFAS/g. This value is higher than that of polymer adsorbent 1–2 log magnitude and lower than the capacity of anion exchange resin and synthesized adsorbent 1 log magnitude. Their equilibrium time also varies widely. The advanced materials reach equilibrium in only a few hours while the traditional adsorbent needs a few days to reach equilibrium.

Recently, amine-based sorbent has emerged as a novel synthesized material which performs effectively in PFAS remediation. This sorbent contains a nitrogen compound and theoretically carries an amide group in its structure. It can be either in organic form (e.g., chitosan, poly-aniline) or a metal organic framework. The amide group is highly tunable, hydrophobic and charged. This amide group can appear in primary, secondary and tertiary structures and especially, possess a charged quaternary ammonium group. Thanks to those characteristics, the amine-based sorbent displays higher affinity for PFAS sorption compared to carbonaceous and nonionic resin. For example, poly(ethylenimine)-functionalized cellulose and amine-functionalized covalent organic framework can remediate PFAS-impacted water up to 1000 ng/L [91,92]. Amine-based sorbent also has superior pore volume - up to 1000 m²/g and pore width of 2.6 nm - which enables PFAS diffusion to be done effectively. The reaction time is also rapid and only takes some minutes to do [92].

In terms of mechanism, the adsorbents adsorb PFAS via hydrophobicity, ligand exchange, electrostatic interaction, formation of hydrogen bond and the nature of PFAS [107]. Hydrophobic and electrostatic interaction between PFAS anion and positive-charged contribute decisively to PFAS adsorption. This can increase the partitioning value (K_d) of PFAS-adsorbent to 10⁸ L/kg [107]. The regular K_d values of several carbonaceous adsorbents stay below 10⁵ L/kg [93]. However, its success depends on the origin of the materials, such as activated

Table 8

Adsorption capacity of selected adsorbents (mg/g).

Adsorbent	Carbonaceous material	Polymer material	Anion exchange resin	Synthesized material	
PFCA	PFBA (C4)	-	635.69	363.86	
	PFPeA (C5)	-	-	-	
	PFHxA (C6)	0.095 – 235.54	-	37.68 – 1089.76	376.86 - 9203
	PFHpA (C7)	0.031 – 65.53	-	192.95	-
	PFOA (C8)	0.111 – 426.49	0.097 – 34	331.25 – 1436.82	2.32 - 1283.61
PFSA	PFBS (C4)	51.01	-	1023.32	141.04
	PFHxS (C6)	0.121	-	1364.37	224
	PFOS (C8)	0.345 – 712	0.1 – 1455.52	210.05 – 2750.7	17.2 - 58472
Replacement PFAS	GenX (C3)	-	-	-	40 - 200
	F-53B (C8)	1.059	-	2396.84	-
Equilibrium time	Several days	20 min	48 – 96 h	< 2h	
References	[3,93–101,[14],	[14,4,102,103,92],	[14,5,104],	[14,6,105,106,91,90],	

carbon felt [107]. The activated carbon felt is fabricated from the synthetic rayon-based viscose fiber to maximize the hydrophobic interaction and electrostatic attraction towards PFAS. Another important factor for PFAS adsorption is pore size distribution. The most effective pore size is the 1–2 nm range which correlates with PFAS adsorption efficiency. In turn, a pore size of less than 1 nm does not show a correlation [107].

In practice, PFAS present in water and wastewater matrix together with several interferences such as dissolved organic matter, humic acid and minerals [108]. Those interferences also compete PFAS adsorption by dominating the adsorption sites (Fig. 8b). On the surface of adsorbent, interferences create a negative charge and initiate electrostatic repulsion between PFAS and the adsorbent [108]. Thus, in a sophisticated water matrix, the PFAS adsorption capacity is reduced. For instance, in a surface water body, biochar adsorbs 41.2–41.3 mg PFOA/g whereas in wastewater, it reduces capacity, ranging between 27.7–31.7 mg PFOA/g [109].

The adsorption performance is also largely governed by the origin of the PFAS compound itself. Short chain and long chain PFAS are adsorbed in different ways, due to the structure and hydrophobicity of the PFAS compound. The short chain PFAS are more hydrophilic and tend to be less adsorbed than long chain compounds. Also, the sulfonate group is adsorbed less than the carboxyl group. The steric hindrance of the sulfonated group (diameter 3.748 Å) is beyond the value of the carboxyl group (2.165 Å) resulting in slower adsorption. In practice, no PFAS compound exists alone, but they co-present in a matrix. The adsorption capacity of the mixture diminishes compared to the single sorption [5]. The order of equilibrium is as follows: PFOS > PFBS >

PFOA > PFBA [5]. This means the carboxyl group is adsorbed before the sulfonamide group.

Apart from the effect of the functional group, the C–F chain length is another reason for this. When increasing PFAS concentration, the long chain PFAS can block the pores of adsorbents and prevent the adsorption and diffusion of other PFAS compounds [106]. They can also replace the adsorption site of short chain PFAS and desorb them (Fig. 8b). For example, PFOS can form the micelle and semi-micelle and hinder the adsorption process. This also explains why the equilibrium time of PFOS is the highest [5]. Similarly, the breakthrough time of PFAS in the column adsorption correlates to the chain length and the functional group. Several granular activated carbon types absorb short chain PFAS ineffectively, indicating that the adsorption of short chain PFAS is independent of activated carbon types. Thus, an advanced adsorbent for short chain PFAS remediation is needed [6].

Most sorption studies are restricted to batch conditions when evaluating the sorption capacity and kinetics of sorbents. Xiao et al. [93] did a sorption study of 30 PFAS compounds in batch condition and modelling; however, column studies using real groundwater are still lacking. For full-scale purposes, more studies that employ a continuous experiment in column reactor are necessary [94,110]. Column experiment is the closest to the real-life situation, through breakthrough profiles, when sorption is affected by engineering factors such as media bed depth, sorbent size, contact time and fluid conditions. Park et al.

[94] experimented using a rapid small-scale column testing with nine PFAS compounds; they found that half breakthrough bed volume can predict PFAS sorption capacity. It is important since desorption can occur and also for designing an appropriate full-scale column. Longer

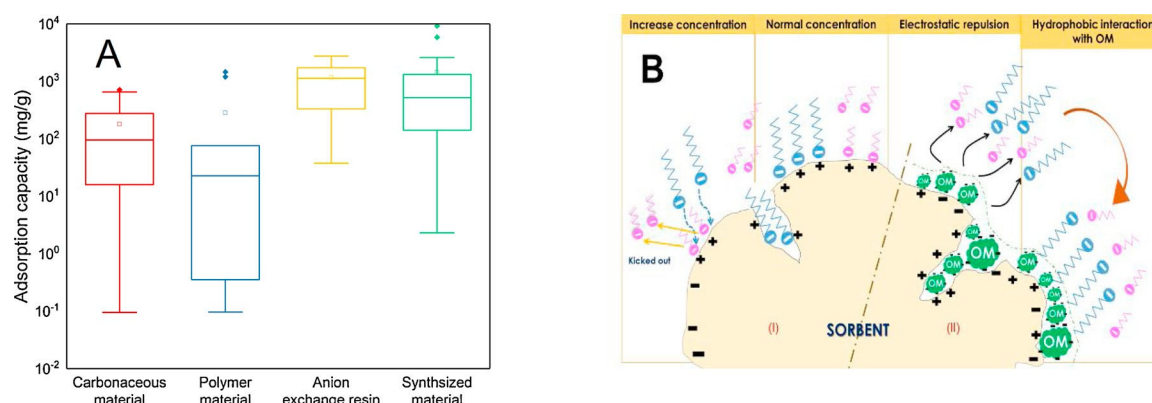


Fig. 8. Adsorption capacity of selected sorbents (A) and mechanism of competitive adsorption of long chain, short chain PFAS and organic matter (B). (I) In a normal concentration, long chain and short chain PFAS adsorb on the adsorbent, but when the PFAS concentration increases, the long chain kicks off the short chain. (II) Organic matter dominates sorption sites on the sorbent's surface and creates electrostatic interaction with the anion head of PFAS. Organic matter attracts the hydrophobic tail of long chain PFAS.

bed contact time results in steeper breakthrough profiles of PFAS. The desorption of short chain PFAS is more severe in longer bed contact time [94]. In a column study, the effect of dissolved organic matter demands special attention because it competes with the transport and sorption of PFAS in the column [110]. By examining UV 254 values of the effluent, Park et al. [94] discovered that the organic matter had reached breakthrough earlier than the PFAS. In other words, organic matter has dominated the sorbent site and pore volume, then phased out the PFAS from the sorbent. It consequently reduced PFAS sorption efficiency.

5.1.2. Stabilization and solidification

The stabilization and solidification (s/s) approaches seek to capture and immobilize PFAS in solid state, but not to degrade them. This technology is particularly feasible for soil, sediment and sludge polluted with PFAS. The current s/s technology deals with PFAS leakage and to manage this issue, additives have been introduced to reduce PFAS leaching. Various additives have been tested including powder activated carbon, Rembind®, pulverized zeolite, chitosan, hydrotalcite, bentonite, and calcium chloride. The powder activated carbon and Rembind® can decrease 70–94 % leaching of 13/14 PFAS compounds [111]. PFOS is stabilized by as much as 99.9 %. The additives have increased the sorption and partitioning of PFAS in solid phase so they reduce the leachate. Thus, less PFAS-contained wastewater is generated.

5.1.3. Membrane technology

Membrane is the typical process of PFAS separation and concentration. This technology is an advanced one in the context of PFAS remediation which rejects high-loading PFAS, up to 1000 mg/L, at extreme efficiency, higher than 99 % [112–115]. The primary mechanism whereby the membrane eliminates PFAS is based on size-exclusion. There are other mechanisms involved in the remediation process: electrostatic repulsion, diffusion and cake layer filtration [112]. The proper membrane type that works for PFAS rejection is nanofiltration and reverse osmosis. Compared to reverse osmosis, nanofiltration is less expensive and is favorable for PFAS removal, especially the NF270 [46].

The nanofiltration NF270 can reject 99 % PFAS and even some PFAS compounds less than 270 Da, such as short chain PFAS [112]. For particular compounds such as PFHxA, the NF270 can reject 96.6–99.4 % PFAS [113]. The PFHxA concentration in the retentate reaches 870 mg/L. The drawback of the membrane is that the concentrate has to be handled by a destructive technology, such as AOP [113]. In this case, electro-oxidation is applied and reduces PFHxA down to 8–21 mg/L. Another study has noted that NF270 can deal with PFAS in ground-water that is highly saline [114].

Membrane is efficient in PFAS remediation but it does suffer from a major technical issue: fouling. This is caused by humic acid and inorganic ions in the groundwater which can compromise PFAS rejection efficiency. It can be fixed by adjusting the flux [116]. Consequently, the membrane can reject 93 % PFAS load, 99 % of NaCl and MgSO₄. The concentration of long chain PFAS in the permeate is below the detection limit but the short chain PFBS and PFBA still present at 10 ng/L. Notably, the fouling membrane does not affect short chain PFAS given that the fouling layer has contributed to short chain PFAS rejection.

Another rejection mechanism of membrane relies on the aggregation of PFAS. PFOA can form a micelle and increase the rejection rate consequently [117]. Although this NF270 is efficient in rejecting long chain PFAS, it experiences difficulty with uncharged compounds. For example, NF270 only retains 42 % of FOSA, especially at pH below 3. The explanation was the charged compound adsorbs quickly to the membrane surface while the uncharged FOSA does not [116]. This is due to the poor electrostatic repulsion of FOSA towards the negatively charged NF membrane. The solution increases pH to 10 which can reject 99 % FOSA.

The membrane method has been employed for PFAS remediation in full-scale settings [46]. For instance, two water treatment plants in the US used RO membrane at capacity 12 gfd receiving recovery 80–85 %. However, as previously mentioned, it is a costly process but NF270 is recommended in this case.

5.2. Destruction technologies

5.2.1. Advanced oxidation process

The AOP can generate hydroxyl radical groups (%OH) which create a strong oxidation state for PFAS degradation. The currently reported AOP include ozonation, catalyst-supported UV, electro-chemical oxidation and air fractionation. The general mechanism of PFAS degradation by AOP is the cleavage of %OH using two possible pathways. The first one is the replacement of H on the C–F bonding. The second route is the cleavage of %OH on the backbone of the PFAS chain, breaking them up into smaller fractions [31,118]. This route includes the following progression: decarboxylation–hydroxylation–elimination–hydrolysis.

Those technologies can either stand alone or be combined in the WWTPs. For example, gallium oxide and peroxymonosulfate catalysts are irradiated by UV (185 and 254 nm) for PFOA degradation. Within 60–90 min, the system degrades 100 % PFOA and can deal with an initial concentration of 50 ng/L to 50 mg/L [9]. PFOA is degraded to PFHxA, PFHpA and PFPeA. This system is efficient for real wastewater application having similar PFOA degradation efficiency outcomes. Another catalyst for UV application is sulfite. This technology has been tested with chrome plating wastewater. This wastewater contains substantial F-53B, a PFAS replacement compound. The results show that F53-B can be degraded in a period lasting from 1 to 60 min. The UV, ozonation and air fractionation are combined for PFAS removal and the integrated system degraded 87 % PFAS whereas the single technology recorded 16.8–73 %. The involvement of air fractionation conditions in PFSA degradation is because gas bubbles have more affinity for the PFSA group, rather than PFCA [119]. With reference to electrochemical oxidation, 90 % of PFOA and PFOS are degraded within 10–30 min depending on current (10–15 mA/cm²) [120].

The AOP can degrade PFAS in a short period of time; however, AOP still cannot deal efficiently with short chain PFAS. For instance, electrochemical oxidation needs 8 h to degrade 50 % short chain PFBA. It should be noted that the feasibility of electrochemical oxidation in full-scale studies is still limited.

5.2.2. Plasma

Plasma is an advanced treatment process aiming to breakdown PFAS extensively. It is driven by high voltage. Here, the reactive oxygen species such as %OH, %O, H%, HO₂%, O₂%, H₂, O₂, H₂O₂ and aqueous electrons (e_{-aq}) are generated. It can degrade 90 % PFOA at a concentration of 8.3 mg/L in 60 min in laboratory conditions [8]. Plasma has been tested in pilot scale studies to evaluate the feasibility. The pilot plasma reactor can degrade a range of PFAS compounds from 1 to 50 min at a concentration of 1.4 mg/L [121]. After plasma treatment, a variety of products can be detected: PFHpA, PFHxA, PFPeA, PFBA for PFOA, and PFHxS and PFBS for PFOS. Those products are continually defluorinated to non-fluoride fractions or ions: CF₃COOH, CH₃COOH, HCOOH, F⁻, and SO²⁻. The details of degradation pathways are described elsewhere [8].

The defluorination mechanism of plasma technology is similar to the AOP process but more intensive. Plasma technology produces substantial non-fluoride products and gaseous by-products (C₄F₈, C₅F₁₀, C₆F₁₂, C₇F₁₄, and C₈F₁₆) compared to AOP. In addition, plasma can deal with PFAS precursors. This explains why plasma is a profound PFAS remediation option. By interacting with the precursors, plasma can degrade precursors within 1 min to below the US EPA standard given the total of oxidizable precursors are recorded at 10² to 10⁵ ng/L [121]. The major concern of this technology is the high voltage required, up to

30 kV. This can mean significant costs for full-scale treatment and safety issues. There is as yet no literature on the economic feasibility of this technology.

5.2.3. Thermal destruction

Thermal destruction technology refers to using high temperature to incinerate and defluorinate PFAS. This technology needs the support of separation technology in advance because it is unable to deal with PFAS in liquid matrix. Sorption and membrane separation are essential to separate PFAS from water for thermal technology. High temperature, up to 700 °C, in a steamed atmosphere or N₂ is the requirements to mineralize PFAS. In practice, PFAS are incinerated with additives (e.g., Na⁺, Ca²⁺) to increase mineralization efficiency [122,123]. Watanabe et al. [122] studied the recovery rate of mineralized fluorine including PFOA, PFHxA and PFOS. At 700 °C, those substances were recovered at 30 %, 46 % and 72 %, respectively. The recovery rates increased to 51 %, 74 % and 70 %, respectively, while PFAS were adsorbed in granular activated carbon beforehand. Additional NaOH increased the recovery rates to 74 %, 91 % and 90 %, respectively. Wang et al. [123] investigated if Ca²⁺ was an effective additive for PFAS mineralization from 300 °C to 900 °C. Among three forms of Ca²⁺ salts (i.e., CaO, CaCO₃ and Ca(OH)₂), Ca(OH)₂ delivered the distinctive performance. At 300 °C, PFAS could not react with three additives, yet at 350 °C, only Ca(OH)₂ started forming crystal CaF₂. The mineralization efficiency rose to 60, made possible by the hydrogen atoms in Ca(OH)₂ molecules participating in hydrogen-defluorination reaction. After increasing the temperature, CaO turned into Ca(OH)₂ and repeated the same reaction. Ca(OH)₂ and CaO also possessed a face-centered cubic crystal structure which gave more opportunity for PFAS to make contact. Thus, they were better than CaCO₃ for defluorination. Although incineration can defluorinate effectively, there is one important drawback. During the incineration process, toxic gases were generated, such as dioxin, furan and fluorocarbon emissions (%CF, %CF₃, %C₂F₄ and %C₃F₅). This issue needed to be investigated and resolved in future studies.

5.2.4. Sonochemical degradation

Sonochemical is another option for PFAS destruction using sono-wave as main driven force. Through soundwave, PFAS are remediated at bubble/water interface. Bubbles which were created by soundwave start to enlarge until they reach the stage of quasi-adiabatic compression and collapse afterwards. This process releases heat up to 5000 K and high pressure. The heat conditions an environment for the pyrolysis of PFAS which adhere at the interfacial area of cavitation bubble. The pyrolysis process also generates free radical groups, such as %H and %OH. Then the free radical groups become involved in the PFAS remediation simultaneously with pyrolysis.

Efficiency of the sonochemical process is greatly dictated by power and frequency used. Additives are combined in the sonochemical process to improve the overall efficiency. The recommended frequency ranges from 202 kHz to 1000 kHz coupling power of 12 kW [124,125]. Gole et al. [124] investigated that, at a frequency 500 kHz–1000 kHz and power 12 kW, the sonochemical process could decompose 90.5 % PFSA, 26.6 % PFCA and 38.4 % FTS. Likewise, Campbell and Hoffmann [125] found that PFOS and PFOA were degraded at 12 % and 23 %, respectively. The degradation rate constants rose proportionally with power density. The single sonochemical process occurred in a rather long duration (e.g., 13 h). Thus, additives were integrated into the remediation process. For example, sulfate iron could assist the sonochemical process for degrading PFOA completely in 90 min [126]. Lee et al. [127] used periodate to increase PFOA remediation efficiency. At 45 mM periodate, PFOA was degraded 96.5 % and defluorination efficiency reached more than 95.7 % in 120 min. Thanks to the additives, experimental duration was reduced 6- to 9-fold. Other engineering factors also generate certain effects in the sonochemical process. This technology prefers low pH thanks to acid-catalyzation. Dissolved oxygen can diminish the decomposition of PFAS since it reduces free

radical groups present in the water matrix. Energy cost of sonochemical process ranges from \$0.015/L to \$0.019/L [124].

5.2.5. Biodegradation process

For years, biotechnology was believed to be inefficient in PFAS remediation. Using PFAS as the sole carbon source for cell build up is challenging for microorganisms. The energy demand for breaking up the C–F bonding is higher than the ability of microorganisms. Recently, some bacterial strains in anammox and anaerobic conditions have emerged as feasible to transform and degrade PFAS [63,77,128,129]. Most of those bioprocesses are linked to the oxidation-reduction mechanism using iron and sulfate. For example, *Acidimicrobium* sp. can degrade 60 % of PFOA and PFOS at an initial concentration of 0.1–100 mg/L after 100 d. The process is conducted in the presence of ferric iron reducing to ferrous form and using H₂ and ammonia as the electron donor [129]. The electron acceptors serve to defluorinate PFOA and PFOS. The detected terminal products are PFBA, PFPeA, PFHxA, PFHpA for PFOA and PFBA and PFBS for PFOS. This proves that

PFOA and PFOS are broken up into smaller fractions. Anaerobic and aerobic microorganisms are also feasible for PFAS remediation in particular conditions. The anaerobic microorganism in sulfate-reducing conditions can biotransform the precursor 6:2 FtTAoS to 6:2 FtTP [77]. They can transform 75 % of 6:2 FtTAoS in 270 d. The possible pathway is the transformation of 6:2 FtTPIAA via an alanine conjugation after desulfonation and decarboxylation [77]. Likewise, in aerobic conditions, the 6:2 FtTAoS are transformed to PFCA and 6:2 FtS. However, the transformation pathway in anaerobic and aerobic conditions are different because the 6:2 FtTP in anaerobic environment does not experience any further degradation like the aerobic one.

It is evident that aerobic and anaerobic microorganisms take more time than the other technologies for PFAS remediation. At least 100 d are required to phase out 70–80 % of initial PFAS. This can be a technical challenge for practical applications. Recently, some extracted enzymes have proven their ability to remediate PFAS at a fast pace. For example, the protein of *Cannabis sativa* L. showed its feasibility for remediating PFAS [63]. It can degrade 98 % of PFOS and PFHxS in only 1 h. This remediation time is significantly less than those microorganisms. The *Cannabis sativa* L. protein possesses a specific mechanism for PFAS degradation. It can change the secondary structure by increasing β -turns and decreasing random coils, α -helix and β -sheets. This feature can assist the hemp protein adsorb PFAS at the hydrophobic site of protein and the side chain of amino acid [63]. Based on a rapid (1 h) and efficient (98 %) process, this suggests it can be applied for PFAS remediation through the use of extracted protein. Some studies have also reported the impact of PFAS on enzyme systems of microorganism (e.g., sucrose and urease) [128,130].

5.2.6. Mechanism of PFAS defluorination by destruction technologies

Defluorination of PFAS by destruction technologies, either from AOP, plasma, thermal, sonochemical or bioprocess, can be undertaken through two main mechanisms: (i) H/F exchange and (ii) chain shortening. These two mechanisms are subjected to both PFCA, PFSA and precursors but their degradation pathways differ in a small way due to the head groups. The details of PFAS destruction are explained in more detail below.

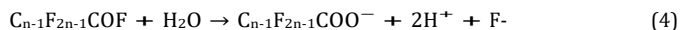
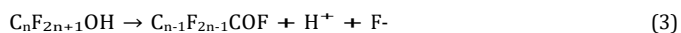
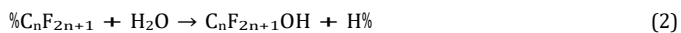
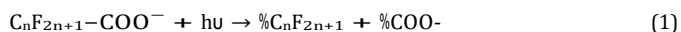
5.2.6.1. PFCA. The degradation of PFCA can be described in four sequential steps: decarboxylation (Eq. 1), hydroxylation (Eq. 2), elimination (Eq. 3) and hydrolysis (Eq. 4) [10]. PFCA possesses a carboxyl group which is highly inductive. The CF₂ moiety linking to the carboxyl group demonstrates high reactivity which is determined via density functional theory calculation. The α position of this CF₂ moiety is encountered by e⁻, and the defluorination process happens. C–F bond is stretched while F⁻ is replaced by H⁻ and H/F exchange is finished in that way. At the same time the carboxyl group is cleaved. The structure of by-products can be written as C_{n-1}F_{2n-1}-CH₂-COOH.

Table 9

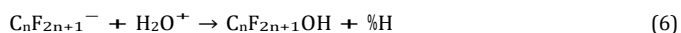
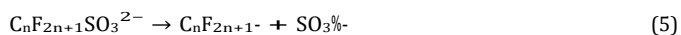
Advantages and disadvantages of PFAS remediation technologies.

Categories	Technology	Application	<i>Ex situ/In situ</i>	Major factors	Advantages	Disadvantages	Applied compounds	Applied water matrix
Separation - Concentration	Adsorption	<ul style="list-style-type: none"> ● Full scale (drinking water treatment plant, treatment train) 	<ul style="list-style-type: none"> ● <i>Ex situ</i> 	<ul style="list-style-type: none"> ● Sorbent dose ● Retention time ● Nature of sorbent 	<ul style="list-style-type: none"> ● Phase out PFAS from aqueous phase ● Low on-site disturbance ● Regeneration ● Low cost (depend on sorbents) 	<ul style="list-style-type: none"> ● Need post treatment by destructive technology ● Impotent to short chain and precursor PFAS ● Competition of co-contaminant ● Time consuming 	<ul style="list-style-type: none"> ● Efficient for long chain PFAS 	<ul style="list-style-type: none"> ● Drinking water ● Ground water (not impacted by AFFF)
	Stabilization and solidification	<ul style="list-style-type: none"> ● Full scale ● Commercial reagents: RemBind, MatCare 	<ul style="list-style-type: none"> ● <i>Ex situ</i> ● <i>In situ</i> 	<ul style="list-style-type: none"> ● Reagent dose ● Additive (activated carbon, alginate) 	<ul style="list-style-type: none"> ● Easy O&M ● Reduce PFAS leaching ● Cheap s/s reagents 	<ul style="list-style-type: none"> ● Longevity of solid phase is unclear, depending on environmental conditions. ● Interference by co-contaminants 	<ul style="list-style-type: none"> ● Applicable for short chain PFAS ● Some commercial reagents work only for PFOS (i.e. MatCare) 	<ul style="list-style-type: none"> ● Sorbent used for those water matrix
	Membrane process	<ul style="list-style-type: none"> ● Lab scale to full scale (NF and RO) 	<ul style="list-style-type: none"> ● <i>In situ</i> (drinking water treatment plant) 	<ul style="list-style-type: none"> ● Type of membrane ● Fouling control ● Flux 	<ul style="list-style-type: none"> ● NF effective for short and long chain PFAS 	<ul style="list-style-type: none"> ● Need post treatment by destructive technology ● Reject water needs to be treated and disposed ● Inefficient for precursor. 	<ul style="list-style-type: none"> ● Most long chain and short chain PFAS 	<ul style="list-style-type: none"> ● Wastewater ● Ground water or water matrix related to AFFF
Destruction	Advanced Oxidation Processes	<ul style="list-style-type: none"> ● Lab scale 	<ul style="list-style-type: none"> ● Can be <i>in situ</i> or <i>ex situ</i> 	<ul style="list-style-type: none"> ● Type of catalyst ● Dose of catalyst 	<ul style="list-style-type: none"> ● Applicable for gases, liquids and solids ● Destructive 	<ul style="list-style-type: none"> ● Unclear degradation pathway of PFAS ● Costly 	<ul style="list-style-type: none"> ● PFOA and PFOS 	<ul style="list-style-type: none"> ● Ground water related to Wastewater ● AFFF
	Plasma	<ul style="list-style-type: none"> ● Pilot scale 	<ul style="list-style-type: none"> ● <i>Ex situ</i> 	<ul style="list-style-type: none"> ● Used voltage ● Inert gas 	<ul style="list-style-type: none"> ● Destructive technology ● Non-fluorine by-products detected 	<ul style="list-style-type: none"> ● High voltage (30kV) ● Costly 	<ul style="list-style-type: none"> ● Several PFAS 	<ul style="list-style-type: none"> ● Concentrate of membrane ● Wastewater ● Ground water related to AFFF
	Thermal	<ul style="list-style-type: none"> ● Pilot 	<ul style="list-style-type: none"> ● <i>Ex situ</i> 	<ul style="list-style-type: none"> ● Used high temperature ● Steamed atmosphere/N2 	<ul style="list-style-type: none"> ● Destructive technology ● Non-fluorine and gaseous byproducts detected 	<ul style="list-style-type: none"> ● Energy cost ● Toxic gas generation 	<ul style="list-style-type: none"> ● Several PFAS 	<ul style="list-style-type: none"> ● Need pretreatment technology to separate PFAS from water
	Sonochemical	<ul style="list-style-type: none"> ● Lab scale to pilot 	<ul style="list-style-type: none"> ● <i>In situ</i> 	<ul style="list-style-type: none"> ● Sonowave 	<ul style="list-style-type: none"> ● Destructive technology ● Non-fluorine by-products detected 	<ul style="list-style-type: none"> ● Need additive to shorten treatment duration 	<ul style="list-style-type: none"> ● Several PFAS 	<ul style="list-style-type: none"> ● AFFF-impacted water
	Bioprocess	<ul style="list-style-type: none"> ● Lab scale 	<ul style="list-style-type: none"> ● Unknown 	<ul style="list-style-type: none"> ● Strain of microorganism ● Type of protein 	<ul style="list-style-type: none"> ● Unclear if the process can be destructive ● Work for liquid 	<ul style="list-style-type: none"> ● phase and potential for solid phase 	<ul style="list-style-type: none"> ● Time consuming, up to 100 d ● Not work for short chain PFAS ● Unclear mechanism 	

The CH₂ moiety can increase the recalcitrance of by-products. PFCA can also be cut at CeC bonding and the shorter chain PFCA is formed. Similar degradation mechanisms happen to the shorter chain PFCA. However, as PFCA become shorter, it is less likely to be degraded.



5.2.6.2. *PFSA*. The degradation pathways of PFSA differ from PFCA due to the sulfonic head group. The head group is cleaved and two groups of by-products are identified: (i) short chain and poly fluorinated PFSA; and (ii) short chain and poly fluorinated PFCA. PFSA is degraded in three steps: desulfonation (Eq. 5), H/F exchange and chain shortening (Eq. 6). The desulfonation refers to the cleavage of C-S bond and forms a carboxyl head group. For this reason, by-products of PFSA include PFCA compounds. The C-S bond carries lower energy (272 kJ/mol) than the CeC bond (346 kJ/mol). Also, the distance of C and S atoms of PFSA is 4.4 Å while the one of C and C atoms of PFCA is 3 times less (1.5 Å) [10]. Hence, the sulfonic head group can be easily replaced. Meanwhile, H/F exchange occurs and creates poly fluorinated compounds. PFSA can also be broken by CeC scissor. According to density functional theory calculation, CeC bonds of C4 to C8 occupy the lowest molecular orbitals. They are the target for e- and heat to strike on and create shorter chain PFAS.



5.2.6.3. *Fluorotelomer precursors*. The fluorotelomer precursors possess -CH₂CH₂- moiety which is recalcitrant for breaking down [118]. Thus, fluorotelomer precursors are not readily degradable compared to PFCA and PFSA which have the same chain length. Only the fluorotelomer possessing carbon chain length more than six has been identified with by-products. The degradation mechanism is similar to PFCA. H/F exchange occurs in C-F bonds at the middle of the chain. It should be noted that chain shortening happens and generates shorter chain PFCA [118].

5.3. Advantages and disadvantages of remediation technologies

PFAS can be remediated via some options as those reviewed above. Each technology has its own advantages but also certain drawbacks. Understanding the origins, usefulness and current status of those technologies is necessary. The advantages and disadvantages of the above PFAS remediation processes are summarized in Table 9. Information here is useful for future research for identifying gaps in our knowledge and seeking solutions. For all the technologies, it is critical

for heading to full scale, onsite application and deploying it in the market place. We also recommend a water matrix that is suitable for each technology, based on the knowledge documented in this review. Some commercialized technologies are listed. However, the information on economic feasibility is still very limited because most of the technologies are still quite new.

Based on the advantages and disadvantages of those technologies, we propose an engineering concept for remediating PFAS in WWTPs (Fig. 9). In the anaerobic treatment stage, the microbes *Acidimicrobium* sp. can be employed to reduce PFAS load [129]. Next, in the aerobic treatment, the activated sludge process spiking protein of *Cannabis sativa* L. can be used [63]. Another option is aerobic MBR integrating NF270 [46]. This MBR technology can also combine with the protein of *Cannabis sativa* L to enhance transformation of PFAS precursors. Finally, in the post-treatment stage, the PFAS destruction technology should be applied. Two possible alternatives are plasma and AOP [9,31,118,121]. If the budget is limited, adsorption is an alternative using a cheap adsorbent such as activated carbon felt [107].

6. Conclusion

PFAS are emerging and bioaccumulative pollutants are widespread in water and wastewater bodies. They have raised several challenges for remediation and management in the last few decades. This review has thoroughly discussed and updated several aspects of PFAS in water and wastewater: sources, occurrences, fate, transformation, transportation and remediation. The following key points need to be addressed in the future, and suggest alternatives:

- Legislation and guidelines: The current legislation and guidelines are still limited and mostly deal with PFOA and PFOS. More attention must be paid to precursors and short chain PFAS.
- Occurrence, detection, identification and quantification: short chain PFAS appear in water and wastewater higher than long chain PFAS at least 1 log magnitude. This confirms the current trend of using the short chain PFAS rather than the long chain PFAS. However, we did not exclude the possibility that long chain PFAS are absorbed in biosolid, sediment. The precursors are troublesome in terms of occurrence since they are difficult to analyze. New precursors are increasingly found in water and wastewater which means that large amounts of precursors remain undetected. It requires cutting-edge facilities such as quadruple time of flight, high resolution mass spectrometry and technical expertise. Also, standard chemicals to quantify them are expensive and lacking in market. Most detected precursors belong to the anionic group. Compared to anionic PFAS, the cationic and zwitterionic PFAS are still poorly detected and quantified.
- Remediation of short chain PFAS and precursors: the short chain PFAS are extremely hydrophilic and difficult to remediate. The precursors might present in water matrix at high concentrations, up to hundred mg/L, but are not yet detected. To transform the

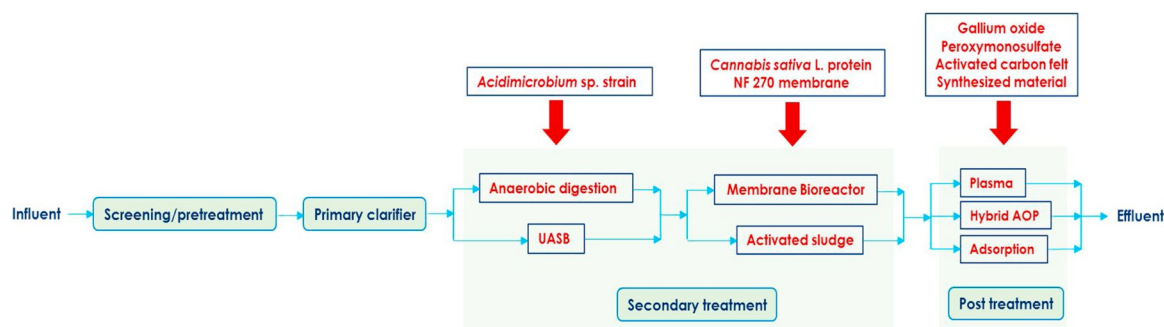


Fig. 9. Proposed wastewater treatment processes for PFAS remediation. The applied resources (e.g., bacteria strain, protein, catalyst) are selected in this review.

precursors to terminal PFAS products, oxidation is needed but this requires large energy consumption. Taken together with short chain PFAS, remediation needs the consecutive steps as follows: separation, concentration and destruction. Ultimately, PFAS need a de-structive technology for defluorination. However, the currently and most widespread technology is adsorption because it is affordable. A destruction technology such as plasma is the most promising one but not yet ready for full scale application.

- **Economic viability and sustainability:** to deploy remediation technology in the market place, a thorough economic assessment of the PFAS remediation technology and management is critically needed. Some key factors for such an economic assessment suggested here are: PFAS matrix, expected outlet and inlet concentration, energy and chemical consumption, expenditure required, epidemiology, sustainable development and effective management.

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.

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