



## Review

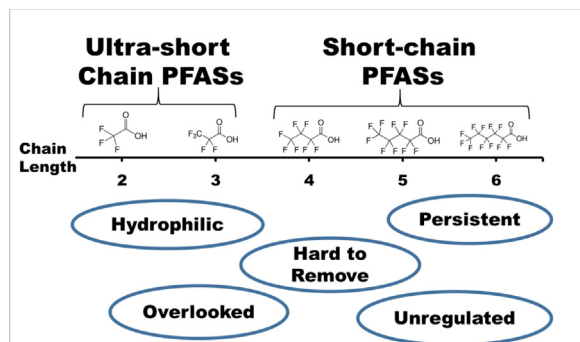
## The overlooked short- and ultrashort-chain poly- and perfluorinated substances: A review

Mohamed Ateia<sup>a</sup>, Amith Maroli<sup>a</sup>, Nishanth Tharayil<sup>b</sup>, Tanju Karanfil<sup>a,\*</sup><sup>a</sup> Department of Environmental Engineering and Earth Science, Clemson University, SC 29634, USA<sup>b</sup> Department of Plant & Environmental Sciences, Clemson University, SC 29634, USA

## HIGHLIGHTS

- Comprehensive surveys of the occurrence of short-chain PFAS are needed.
- Conventional treatment plants fail to remove short-chain PFAS.
- Hybrid sorption systems may be alternatives to remove short-chain PFAS from water.
- Destruction methods are promising but new catalysts and approaches are needed.
- New selective sorbents for short-chain PFAS must be developed.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

## Article history:

Received 5 October 2018

Received in revised form

24 December 2018

Accepted 26 December 2018

Handling Editor: Myrto Petreas

## Keywords:

Poly- and perfluorinated substances

Short-chain

Ultra-short-chain

Occurrence

Treatment technologies

Mobility

## ABSTRACT

Poly- and perfluorinated substances (PFAS) comprise more than 3000 individual compounds; nevertheless, most studies to date have focused mainly on the fate, transport and remediation of long-chain PFAS ( $C > 7$ ). The main objective of this article is to provide the first critical review of the peer-reviewed studies on the analytical methods, occurrence, mobility, and treatment for ultra-short-chain PFAS ( $C = 2-3$ ) and short-chain PFAS ( $C = 4-7$ ). Previous studies frequently detected ultra-short-chain and short-chain PFAS in various types of aqueous environments including seas, oceans, rivers, surface/urban runoffs, drinking waters, groundwaters, rain/snow, and deep polar seas. Besides, the recent regulations and restrictions on the use of long-chain PFAS has resulted in a significant shift in the industry towards short-chain alternatives. However, our understanding of the environmental fate and remediation of these ultra-short-chain and short-chain PFAS is still fragmentary. We have also covered the handful studies involving the removal of ultra-short and short-chain PFAS and identified the future research needs.

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\* Corresponding author.

E-mail address: [tkaranf@clemson.edu](mailto:tkaranf@clemson.edu) (T. Karanfil).

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## Abbreviations

AFFF	Aqueous film-forming foam	PFCA	Perfluorocarboxylic acids
AOF	Absorbable organic fluorine	PFDS	Perfluorodecane sulfonic acid
AOX	Adsorbable Organic Halogens	PFHpS	Perfluoroheptanesulfonic acid
BDD	Boron-doped diamond	PFHxS	Perfluorohexane sulfonic acid
CNT	Carbon nanotubes	PFHxS	Perfluorohexanesulfonic acid
EOX	Extractable Organic Halogens	PFOA	Perfluorooctanoic acid
FASEs	Perfluoroalkyl sulfonamidoethanols	PFOS	Perfluorooctanesulfonic acid
FTOHs	Fluorotelomer alcohols	PFPeS	Perfluoropentanesulfonic acid
GAC	Granular activated carbon	PFSA	Perfluoroalkyl sulfonic acids
HDPE	High-density polyethylene	PIGE	Particle-induced gamma-ray emission
IX	Ion exchange	POPs	Persistent organic pollutants
LC-MS/MS	Liquid chromatography mass spectrometry	PP	Polypropylene
LOD	Limits of detection	QqQ	Quadrupole mass spectrometers
NIX	Non-ion exchange	qTOF-MS	Quadrupole time-of-flight mass spectrometry
NOM	Natural organic matter	SPE	Solid-phase extraction
PAC	Powdered activated carbon	TFA	Trifluoroacetic acid
PFAS	Poly/perfluorinated alkyl substances	TOC	Total Organic Carbon
PFBA	Perfluorobutanoic acid	TONO	Total N-Nitrosamine
		TOP	Total oxidizable precursor

## 1. Background

Poly/perfluorinated alkyl substances (PFAS) and their derivatives represent a large family of anthropogenic organic compounds that since the 1940s have been used in a variety of industries (e.g., chromium plating, aviation hydraulic fluid, and stockpiles of fire-fighting foams) as processing additives and as surfactants since 1940s (Lindstrom et al., 2011; Jian et al., 2017; Wang et al., 2017b, 2017c). The molecular structures of PFAS consist of an aliphatic chain (or multiple chains), in which either all (i.e. per-) or part (i.e. poly-) of the hydrogens are replaced by fluorine such that they contain at least one fluoroalkyl moiety ( $C_nF_{2n+1}$ ) with a terminal functional group (e.g., carboxylates, sulfonates, sulphonamides, phosphonates and alcohols) (Buck et al., 2011). Common consumer products that utilize PFASs include non-stick cookware, specialized garments and textiles, stain repellents, metal plating, and fire-fighting foams. Two main groups of PFAS used in the industry are perfluoroalkyl sulfonic acids (PFSA) and perfluorocarboxylic acids (PFCA) (Rayne and Forest, 2009). In recent years, the occurrence, fate and removal of PFAS in the aquatic environment have been documented as a key emerging issue (Ahrens, 2011; Ahrens and Bundschuh, 2014; Rahman et al., 2014; Land et al., 2015; Banzhaf et al., 2017; Gebbink et al., 2017a).

Due to the persistent and bioaccumulative nature of certain PFAS chemicals (Kannan, 2011),  $C_8$ – $C_{14}$  PFAS and their sodium and ammonium salts were included in the candidate list of regulatory

substances in the EU, and perfluorooctanesulfonic acid (PFOS) has been added to the persistent organic pollutants (POPs) list at the Stockholm Convention on Persistent Organic Pollutants in 2009 (UNEP, 2014). As a result, manufacturers has begun adopting short-chain and ultra-short chain PFAS and other fluorinated alternatives (e.g., perfluoropolyethers) for commercial production (Barzen-Hanson and Field, 2015; Sun et al., 2016; Barzen-Hanson et al., 2017a; Kaboré et al., 2018). However, several challenges characterize the application of short-chain PFAS (Jensen and Warming, 2015; Wang et al., 2015). For instance, the technical performance of short-chain alternates are lower than long-chain PFAS; hence much larger quantities of the short chain PFAS are utilized to obtain a similar performance to long-chain PFAS (Lindstrom et al., 2011). Additionally, several studies have shown that the fluorinated parts of these alternatives are recalcitrant and can form persistent dead-end transformation products (PFCA and PFSA) (Hurley et al., 2004; Lee et al., 2010a; Liou et al., 2010; Butt et al., 2014). Many short-chain PFAS remain unknown/unidentified because they are either proprietary or manufacturing byproducts (Wang et al., 2017c, 2018b). This has resulted in a high uncertainty about the toxicity of short-chain PFASs, especially regarding long-term effects. However, the continuous release of these compounds suggests that adverse effects may occur (Brendel et al., 2018).

PFAS are categorized as ultra-short-chain and short-chain PFAS, based on the number of  $CF_2$  moieties, when they have 2–3 and 4–7 fully fluorinated C-atoms, respectively (Wang et al., 2017c). Despite

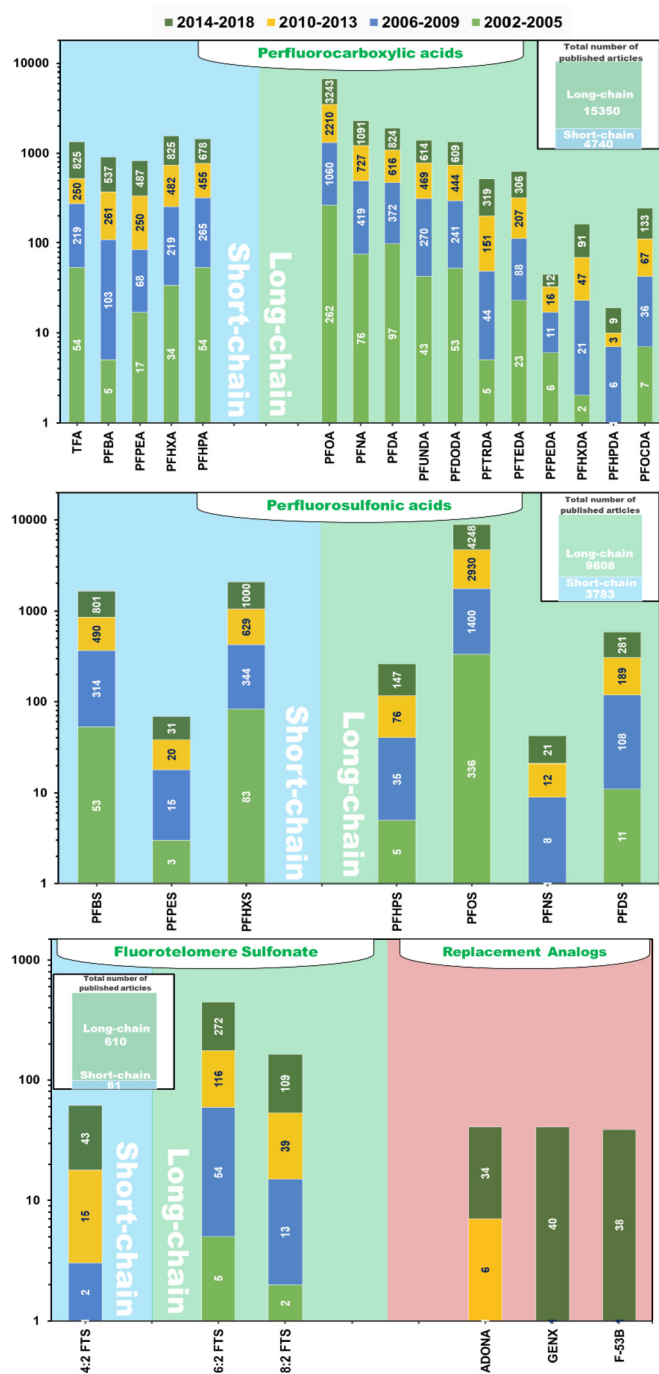


Fig. 1. The number of peer-reviewed studies on PFAS with different chain lengths published between 2002 and 2018. Publications were searched primarily using the Scopus and Google Scholar databases.

a significant shift in the industry towards short-chain alternatives, there are very limited studies on short- and ultra-short PFAS (Fig. 1). The absence of comprehensive knowledge about the environmental fate of ultra-short- and short-chain PFAS underscores the need for further research focusing on novel approaches to remove these compounds from water, as we will discuss in the following sections. To date, there is no review article in literature focusing on ultra-short- and short-chain PFAS. Therefore, the main objectives of this article are to provide a critical review of the published studies that are related to the occurrence, mobility, treatment and analytical methods for ultra-short- and

short-chain PFAS, and to identify the future research needs.

## 2. Measurement methods

To cope with the dynamic industrial production of various PFAS, the researchers need quick adoption of monitoring programs and analytical techniques that should cover those emerging compounds. Research studies on the measurement of PFAS in a variety of matrices (e.g., surface waters, sea waters, landfill leachate) have been conducted over the past decades (Jahnke and Berger, 2009; Banzhaf et al., 2017; Domingo and Nadal, 2017; Kucharzyk et al., 2017; Rappazzo et al., 2017). The improvement in column chemistry, analytical instruments, separation/concentration, and selective and sensitive methods have resulted in the measurement of previously undetected PFAS (Newton et al., 2017; Xiao et al., 2017a). Historically, the inability of achieving precise determination of PFAS from environmental samples have been due to the lack of isotopically labeled standards and limited access to sensitive instruments to detect low/trace concentrations (Trojanowicz and Koc, 2013). It is possible depending upon the method used, to identify and quantify PFAS either as individual species or collectively as total fluoride species. The different methods are described in the succeeding sections.

### 2.1. Important considerations for sample handling and extraction

The structural diversity PFASs in the market as well as the coexisting degradable polymers in the environment possess several technical challenges with the current established analytical methods. Thus, the proper collection, storage and preservation of samples for PFAS analysis is of critical importance to mitigate the losses and contamination can easily occur at any of these stages. When low concentrations of the PFAS are targeted (ng/L or less), great care must be taken to avoid sample contamination. One common method entails pre-cleaning the sampling bottles prior to sampling by rinsing with polar/semi-polar solvents such as de-ionized water, acetone or methanol (González-Barreiro et al., 2006; Skutlarek et al., 2006). Regardless of these careful efforts, traces of PFOA contamination were nonetheless present in USEPA recommended polypropylene (PP) sample bottles (Yamashita et al., 2004). Other studies have shown that water samples stored in PP or high-density polyethylene (HDPE) bottles showed irreversible adsorption of PFAS to the sample container surface, a finding that was more prominent for long-chain PFASs (>C<sub>10</sub>), PFOSA and NETFOSA (Taniyasu et al., 2005; González-Barreiro et al., 2006). In contrast, owing to their high solubility, ultra-short- and short-chain PFAS remain in solution, a finding supported by the absence of their sorption to container surfaces (Pawel et al., 2006). However, solvent-rinsed PP rather than glass bottles must remain the standard for sample collection and storage. Water samples for PFAS analysis are often stored in freezers or refrigerators with such samples kept at room temperature for short-term storage. Acidification of water samples during storage should be avoided as the proton association may lead to adsorption to the sample container (Martin et al., 2004). The storage of biologically active samples such as sewage sludge is also possible as supported by several studies detailing the biotransformation of FTOHs into saturated and unsaturated PFSA that could potentially change the PFAS composition in the sample (Mary Joyce et al., 2004; Wang et al., 2005).

As the PFAS concentrations in the aquatic samples are in ng/L range, which is much lower concentration than some other background analytes, the extraction, and pre-concentration of water samples prior to analysis are necessary to enable detection. Solid-phase extraction (SPE) is the preferred sample extraction procedure for determining PFAS from aqueous matrices is (Ahrens, 2011).

The sorbents most commonly used in SPE are weak anion-exchangers like Oasis WAX (Zacs and Bartkevics, 2016; Gremmel et al., 2017; Yeung et al., 2017b) and Strata-X AW (Munoz et al., 2015; Zacs and Bartkevics, 2016) and polymeric reversed phases or hydrophilic-lipophilic sorbents as Oasis HLB (Wang et al., 2011; Portolés et al., 2015) and Strata-X (Campo et al., 2016). Methanol is used for the elution of PFAS from the reverse-phase sorbent (Gremmel et al., 2017), specifically via  $\text{NH}_4\text{OH}$  (0.1%–2%) in methanol used to elute PFAS from a weak anionic bed to reverse the charge on the sorbent bed (Boiteux et al., 2016; Lin et al., 2016). However, SPE was also identified as a potential source of background contamination, as an evaluation of two different SPE cartridges (OASIS HLB and Sep-Pak C18) shows, which indicated the presence of notable amounts of PFAS contamination (Yamashita et al., 2004). While the Sep-Pak C18 exhibit strikingly high levels of PFOA and PFOS detected, the Oasis HLB cartridges exhibited a background presence of PFOS, PFOA, PFHS, and PFBS, albeit at lower concentrations than that in the Sep-Pak cartridges.

## 2.2. Total organic fluorine (TOF) analysis

Total indices are widely used in modern analytical chemistry in environmental, food, clinical and process analysis, to describe the content of a group of chemical compounds that share similar nature and properties. The commonly used total indices in environmental analysis include Adsorbable Organic Halogens (AOX), Extractable Organic Halogens (EOX), Total N-Nitrosamine (TONO), and Total Organic Carbon (TOC) (Hayer et al., 1996; Kulshrestha et al., 2010).

Advances in manufacturing techniques have resulted in the production (intentional and unintentional) of diverse PFAS compounds with added structural and chemical complexity. However, current analytical techniques can only capture a limited number of these compounds (as discussed in the following sub-sections). Owing to this limitation, the unknown and uncaptured PFAS will not be considered in any health and/or environmental assessments. To cope with this gap, it is vital to make available analytical methods to determine the total organic fluorine by measuring the release of fluorine from organic compounds. Table 2 summarizes the most studied methods for TOF measurements. Among these, the absorbable organic fluorine (AOF) assay relies on an adsorbent to absorb the extractable organic fluorine compounds for purposes of determining the total fluorine through the combustion and detection of fluoride with ion chromatography (Miyake et al., 2007a, 2007b; Wagner et al., 2013). Similarly, particle-induced gamma-ray emission (PIGE) spectroscopy provides the sum parameter of total fluorine abundance (Ritter et al., 2017). The detection limits of AOF and PIGE methods are in the range of  $\mu\text{g/L}$ . Total oxidizable precursor (TOP) assay overcomes the shortcomings that characterize previous methods by lowering the limits of detection to the range of 1–2 ng/L (Houtz and Sedlak, 2012). TOP assay involves the oxidation of all precursors present in the sample to generate PFASA and PFSA to provide rapid and easy assessments of the mass of all PFAS (i.e., precursors and dead-end products) (Houtz et al., 2016). Moreover, an analysis of the sample before and after the oxidative pretreatment allows for a quantitative estimate of the precursor content in the sample.

## 2.3. Individual PFAS speciation analysis

**Gas chromatography (GC):** Early analytical determination of PFAS was carried out using GC coupled to a flame ionization detector (GC-FID) and electron capture detector (GC-ECD) (Belisle and Hagen, 1978). Shafique et al. (2017) have summarized the timeline of reported GC methods for the determination of PFCAs in different matrices. However, the GC analysis of ultra-short- and short-chain

PFAS is poorly understood compared to long chain PFAS ( $C > 7$ ). Of the studies available regarding the GC analysis of short-chain PFAS, chemical derivatization with 2,4-difluoroaniline in the presence of *N,N*-dicyclohexylcarbodiimide has been used to produce 2,4-difluoroanilides of the  $C_2$ – $C_9$  acids (Scott et al., 2006a, 2006c). Consequently, a derivatization step prior to GC analysis is necessary given the low volatility and high polarity of PFAS. Various derivatization procedures have also been devised to convert PFAS into their methyl esters by reactions with diazomethane (Belisle and Hagen, 1980; Henderson et al., 2007), methyl iodide (Moody and Field, 1999), methanolic  $\text{BF}_3$  (Alzaga et al., 2005), and methanol/MTBE/DCA (Fujii et al., 2012). In their studies to determine the presence of perfluorinated organic acids (PFAS,  $C_6$ – $C_{12}$ ) on GC, Dufková et al. (2009) used isobutyl chloroformate (IBCF) to convert the acids into more volatile isobutyl esters, under catalysis by pyridine. In addition to this rapid and simple derivatization procedure, they utilized various extraction matrices (acetonitrile, water, methanol, phosphate buffer) and detectors (GC-EI-MS, GC-ECD). The limits of detection (LOD) which ranged from 0.06 to 1.80 mg/L and 0.030–0.314 mg/L obtained with GC-ECD and GC-EI-MS, respectively, were dependent upon the matrix and the detector used (Shafique et al., 2017).

**Liquid chromatography mass spectrometry (LC-MS/MS):** High-performance liquid chromatography (HPLC) coupled to triple quadrupole mass spectrometers (QqQ) is currently the most preferred and extensively employed analytical method for PFAS quantitation. Triple quadrupole tandem MS (MS/MS) is often used because of its availability across analytical laboratories as well as its ability providing high sensitivity and linearity in a wide range for PFAS determination (Taniyasu et al., 2005). It is now possible to detect PFAS at a pg/L level with newer analytical instruments coupled with the commercial availability of native and mass labeled standards (Vestergren et al., 2012; Filipovic et al., 2015). However, the possible presence of multiple PFOS isomers and different PFAS subgroups in environmental samples has more thoroughly elucidated that PFAS chemistry (Benskin et al., 2010). Although quantitatively robust for trace analysis, conventional QqQ is limited by low resolution in the discrimination of isobaric species. In contrast, high-resolution mass spectrometry (e.g. quadrupole time-of-flight mass spectrometry (qTOF-MS) and Orbitraps) provides a high resolving power, selectivity and mass accuracy necessary for the discovery of novel PFAS for which no reference standards are yet available (Berger et al., 2004; Riddell et al., 2009; Kaufmann et al., 2010; Fang et al., 2016; Zhang et al., 2018). Further, the non-targeted profiling of various biological and environmental sources has yielded new short-chain homologs and novel classes of PFAS (Lisa and Scott, 2014; Rotander et al., 2015). In a study of screening PFAS in blood serum of firefighters exposed to aqueous film-forming foam (AFFF), several short-chain PFAS such as perfluorohexanesulfonic acid (PFHxS), perfluoropentanesulfonic acid (PFPeS), perfluoroheptanesulfonic acid (PFHpS), ether-PFHxS and Cl-PFHxS were detected (Rotander et al., 2015). With the aid of qTOF, previously unidentified classes of novel anionic, zwitterionic, and cationic PFASs were tentatively identified, with more than 3/4th of the novel classes identified comprising short-chain and fluorotelomer-derived analogs (Barzen-Hanson et al., 2017b). Other non-conventional analytical methods using ultra performance convergence chromatography (UPC<sup>2</sup>) coupled to a tandem mass spectrometer have also yielded measurements of the PFAS including the ultrashort-chain ( $C_2$ – $C_3$ ) forms (Yeung et al., 2017b).

Despite the advancements in these instrumentation techniques, analytical separation and detection of short-chain PFAS compounds remains a challenge. These polar short-chain compounds are less compatible for the simultaneous extraction and quantitation along with the long-chain PFAS analytes in routine reverse-phased

**Table 1**  
Occurrence of PFAS in different types of aqueous environments.

Occurrence	Region	Sampling period	Detected compounds	Category	Mean Concentration (pg/L)	Ref.			
Sea/Ocean	Arctic Ocean	1998	TFA	Ultra-shortchain	8000–181000	Scott et al. (2005)			
	Atlantic Ocean	2002	TFA	Ultra-shortchain	17000–200000				
	Pacific Ocean	1999	TFA	Ultra-shortchain	1000–230000				
	Mediterranean Sea	Tokyo Bay, Japan	2005	TFA	Ultra-shortchain	500–50000	Yamashita et al. (2004)		
			2002–2003	PFHS	Short-chain	4900			
				PFOS	Long-chain	19100			
		Shandong peninsula, China	2012	PFOA	Long-chain	173200		Wan et al. (2017)	
				PFBA	Short-chain	2100			
			PFPA	Short-chain	2900				
			PFHxA	Short-chain	9500				
			PFHpA	Short-chain	2900				
			PFOA	Long-chain	35600				
			PFNA	Long-chain	2600				
			PFDA	Long-chain	1900				
			PFUnDA	Long-chain	1600				
			PFDoDA	Long-chain	700				
		PFBS	Short-chain	1100					
		Coastal area of Hong Kong	2002–2004	PFHxS	Long-chain	3400	Yamashita et al. (2005)		
				PFOS	Long-chain	12900			
	PFOA		Long-chain	3100					
	PFOS		Long-chain	1300					
	Coastal area of Korea		2002–2004	PFOA	Long-chain	5700		Yamashita et al. (2005)	
				PFOS	Long-chain	1300			
Rivers	Ganges River, India	2014	PFPA	Short-chain	1475	Sharma et al. (2016)			
			PFHxA	Short-chain	2171				
			PFHpA	Short-chain	1229				
			PFOA	Long-chain	403				
			PFNA	Long-chain	76				
			PFDA	Long-chain	68				
			PFBS	Short-chain	5554				
			PFHxS	Long-chain	120				
			PFOS	Long-chain	507				
			River Elbe, Germany	2007	PFPA		Short-chain	2000	Ahrens et al. (2009)
					PFHxA		Short-chain	3300	
					PFHpA		Short-chain	1600	
					PFOA		Long-chain	6200	
					PFNA		Long-chain	700	
	PFDA	Long-chain			400				
	PFBS	Short-chain			2600				
	PFHxS	Long-chain			1100				
	PFOS	Long-chain			1700				
	Llobregat River, Spain	2010			PFPA	Short-chain	1300	Campo et al. (2015)	
			PFHxA	Short-chain	13500				
			PFHpA	Short-chain	17200				
			PFOA	Long-chain	75300				
			PFNA	Long-chain	30100				
			PFDA	Long-chain	2400				
			PFBS	Short-chain	2200				
			PFHxS	Long-chain	18600				
			PFOS	Long-chain	1720000				
			Lambro River, Italy	2011–2012	PFBA	Short-chain	26000		Castiglioni et al. (2015)
	PFHxA	Short-chain			31000				
	PFHpA	Short-chain			47500				
	PFOA	Long-chain			159000				
	PFNA	Long-chain			86000				
	PFDA	Long-chain			52000				
	PFBS	Short-chain			33000				
	PFHxS	Long-chain			2500				
	PFOS	Long-chain			27000				
	Samondogawa River, Japan	2010–2012			PFBA	Short-chain	9000	Takemine et al. (2014)	
			PFHxA	Short-chain	8000000				
			PFHpA	Short-chain	14000				
			PFOA	Long-chain	180000				
			PFNA	Long-chain	19000				
			PFDA	Long-chain	26000				
			PFBS	Short-chain	2000				
			PFHxS	Long-chain	4200				
			PFOS	Long-chain	48000				
			Hanjiang River, China	2010	PFOA	Long-chain	41000		Wang et al. (2013a)
	PFNA	Long-chain			9500				
PFDA	Long-chain	13200							
PFOS	Long-chain	26100							
River water, the Netherlands	2016	PFBA	Short-chain	5400	Gebbink et al. (2017b)				
		PFPA	Short-chain	4500					



Table 1 (continued)

Occurrence	Region	Sampling period	Detected compounds	Category	Mean Concentration (pg/L)	Ref.	
Rain and Snow Water	Tsukuba City and Kawaguchi City, Japan	2007	PFHxA	Short-chain	6200	Taniyasu et al. (2008)	
			PFHpA	Short-chain	1900		
			PFOA	Long-chain	4700		
			PFNA	Long-chain	710		
			PFDA	Long-chain	410		
			PFBS	Short-chain	22000		
			PFHxS	Long-chain	2100		
			PFOS	Long-chain	3600		
			GenX	Emerging - Long-chain	420000		
			PFOS	Long-chain	500		
			PFDoDA	Long-chain	250		
			PFUnDA	Long-chain	600		
			PFDA	Long-chain	550		
			PFNA	Long-chain	2000		
			PFOA	Long-chain	1100		
			PFHpA	Short-chain	1000		
			PFHxA	Short-chain	700		
			PFPeA	Short-chain	400		
			PFBA	Short-chain	900		
			PFPrA	Ultra-shortchain	9500		
TFA	Ultra-shortchain	65000					
Surface/urban runoffs	Switzerland	1996	TFA	Ultra-shortchain	151000	Berg et al. (2000) (Frank et al., 1995, 1996) (Zehavi and Seiber, 1996; Wujcik et al., 1998, 1999) Von Sydow et al. (2000) Berg et al. (2000) Scott et al. (2006b)	
	Germany	1994–1995	TFA	Ultra-shortchain	200000–700000		
	CA, USA	1995–1997	TFA	Ultra-shortchain	55000–253000		
	Ireland	1996	TFA	Ultra-shortchain	45000		
	Vietnam	1996	TFA	Ultra-shortchain	75000		
	Canada	2002	TFA	Ultra-shortchain	110000		
	Guangzhou, China	2007–2008	PFPrA	Ultra-shortchain	25000		
	New York City, USA	2006	TFA	Ultra-shortchain	225000		
	Tokyo, Japan	2005	PFHpA	Short-chain	900		Murakami et al. (2009)
	PFOA		Long-chain	5100			
PFDA	Long-chain		1050				
PFOS	Long-chain		1700				
PFOA	Long-chain		90000				
Drinking water	Little Hocking, USA	1984–2004	PFNA	Long-chain	43000	Paustenbach et al. (2006)	
	Belpre, USA	2002	PFDA	Long-chain	22000		
	Lubeck, USA	1991–2000	PFUA	Long-chain	44000		
	Tuppers plains, USA	2001–2003	PFDDA	Long-chain	71000		
	Pomeroy, USA	2002	PFOS	Long-chain	50000		
	Arnsberg-Neheim, Germany	2006	PFOA	Long-chain	600000		
	Drinking water, the Netherlands	2016	PFHxA	Short-chain	3100		Gebbink et al. (2017b)
	Ruhr area, Germany	2006	PFOA	Long-chain	4000		
	PFBS		Short-chain	6000			
	GenX		Emerging - Long-chain	3000			
PFPeA	Short-chain		39000				
PFHxA	Short-chain		21000				
Blacktown, Australia	2010	PFOA	Long-chain	110000	Thompson et al. (2011)		
PFBS		Short-chain	12000				
PFOS		Long-chain	9000				
PFPeA		Short-chain	1820				
PFHxA		Short-chain	770				
Groundwater	Ganges River, India	2014	PFHpA	Short-chain	730	Sharma et al. (2016)	
			PFOA	Long-chain	1640		
			PFBS	Short-chain	740		
			PFHxS	Long-chain	3470		
			PFOS	Long-chain	3920		
			PFPA	Short-chain	1475		
			PFHxA	Short-chain	2171		
			PFHpA	Short-chain	1229		
			PFOA	Long-chain	403		
			PFNA	Long-chain	76		
Nevada, USA	1999	PFDA	Long-chain	68	Moody and Field (1999)		
		PFBS	Short-chain	5554			
		PFHxS	Long-chain	120			
		PFOS	Long-chain	507			
		PFHxA	Short-chain	260000000			
			PFOA	Long-chain	75000000		

(continued on next page)

Table 1 (continued)

Occurrence	Region	Sampling period	Detected compounds	Category	Mean Concentration (pg/L)	Ref.
	Military bases, USA	2001–2014	PFNA	Long-chain	315000000	Barzen-Hanson and Field (2015)
			PFETs	Ultra-shortchain	4000000	
			PFPrS	Ultra-shortchain	20000000	
			PFBS	Short-chain	60000000	
AFFF-impacted site, USA	2014–2016		PFHxS	Long-chain	250000000	Xiao et al. (2017b)
			PFOS	Long-chain	100000000	
			PFBA	Short-chain	3700000	
			PFPeA	Short-chain	5200000	
			PFHxA	Short-chain	7600000	
			PFHpA	Short-chain	1500000	
			PFOA	Long-chain	10700000	
			PFBS	Short-chain	1800000	
			PFHxS	Long-chain	21000000	
			PFHpS	Long-chain	1700000	
Military bases, USA	1942–1990		PFOS	Long-chain	33100000	Backe et al. (2013)
			4:2 FtS	Short-chain	7500000	
			6:2 FtS	Short-chain	220000000	
			PFBS	Short-chain	4300000	
			PFHxS	Long-chain	240000000	
			PFHpS	Long-chain	11000000	
			PFOS	Long-chain	78000000	
			PFBA	Short-chain	24000000	
			PFPeA	Short-chain	69000000	
			PFHpA	Short-chain	130000000	
			PFOA	Long-chain	51000000	

analytical methods. Indeed, short-chain compounds most often elute early and exhibit poor peak resolutions (Ruan and Jiang, 2017). Consequently, the efforts to improve their detection, peak resolution and quantitation been the subject of considerable research with trials undertaken on additional stationary phases or separation mechanisms, particularly hydrophilic interaction liquid chromatography (HILIC) and convergence chromatography (Ruan

and Jiang, 2017; Lorenz et al., 2018).

Although HPLC coupled to triple quadrupole (QqQ) is the most widely used analytical instrument for PFAS detection at low LODs, the unit resolution of these mass analyzers shows inadequate selectivity. Therefore a high-resolution MS (HRMS) technique has been used to circumvent this limitation in terms of distinguishing the various PFAS including branched isomers and structural

Table 2  
Methods for total organic fluorine (TOF) measurement.

Method	LOD (ppb)	Concept	Pros	Cons	Ref.
PIGE	2.0–15.0	Isolates organofluorine compounds on solid phase extraction that measures total fluorine.	<ul style="list-style-type: none"> <li>- Rapid screening technique.</li> <li>- Non-destructive.</li> <li>- Quantitative screening.</li> <li>- Provides a measurement of total fluorine.</li> </ul>	<ul style="list-style-type: none"> <li>- No identification of PFAS.</li> <li>- No speciation of PFAS chain length.</li> <li>- Requires nuclear excitation instead of atomic excitation.</li> </ul>	Ritter et al. (2017)
AOF/CIC	1.0	PFAS and other organic constituents are sorbed to an activated carbon matrix that is combusted with the resultant fluoride measured via ion chromatography.	<ul style="list-style-type: none"> <li>- Rapid screening technique.</li> <li>- Captures all PFAS. Provides a measurement of total organic fluorine.</li> </ul>	<ul style="list-style-type: none"> <li>- Total fluorine measurement – not identification.</li> <li>- No speciation of PFAS chain length.</li> <li>- Requires high combustion</li> </ul>	Wagner et al. (2013)
TOP	~0.002	A parallel procedure for the chemical and thermal treatment of aqueous extract. Samples are first analyzed for PFASA, PFSA and any identified precursors. A second aliquot of the sample is then subjected to harsh oxidative conditions. The oxidized sample is then analyzed for the same PFAs and precursors. Expect a A) Reduction of the precursors and B) Increase in concentrations of perfluoroalkyl acids.	<ul style="list-style-type: none"> <li>- Rapid screening technique.</li> <li>- Measures the total mass of precursors to the PFAA.</li> <li>- Sensitivity (ppt) better than PIGE and AOF/CIC.</li> <li>- Readily applicable for aqueous samples.</li> </ul>	<ul style="list-style-type: none"> <li>- Requires prior extraction for non-aqueous samples.</li> <li>- Potential for exhaustion of the oxidant, with incomplete degradation.</li> <li>- Provides a conservative estimate</li> </ul>	Houtz and Sedlak, (2012)
HRMS (TOF/Orbitrap)	~0.001	High-resolution MS distinguishes various PFAS including branched isomers and structural homologs in complex environmental matrices.	<ul style="list-style-type: none"> <li>- Compositional identification.</li> <li>- Identifies multiple precursors via mass ions capture and accurate mass estimation.</li> </ul>	<ul style="list-style-type: none"> <li>- Semi quantitative</li> </ul>	Munoz et al. (2016)

LOD: limit of detection.

homologs in complex environmental matrices (Munoz et al., 2015; Pico et al., 2015; Zacs, 2015; Zacs and Bartkevics, 2016). HRMS is superior to the conventional MS/MS is that it provides accurate mass information for use in deducing the most probable empirical formula for the unknown entity. High-resolution time-of-flight and orbitrap mass analyzers have already been used in the routine monitoring of PFAS in various environmental samples (e.g., surface waters, sea waters, landfill leachate) (Yamashita et al., 2004; Liu et al., 2015; Munoz et al., 2015, 2016; Chiesa et al., 2018). Additionally, unlike other screening assays such as TOP and PIGE, non-targets analysis by HRMS screening provides information on the species identities. HRMS been instrumental in deriving the identification of newly introduced compounds, which have been used as substitute for such regulated compounds as PFOS and PFOA in industrial processes and formulates. However, despite their high mass accuracy and resolution, analyses of PFAS from environmental samples are often characterized by a high background noise and significant interference on the chromatograms, thus rendering them ineffective for quantitative analyses (Zacs, 2015; Cervený et al., 2016; Lin et al., 2016; Lorenza et al., 2018).

### 3. Occurrence and mobility in natural waters

Generally, PFAS exist as anionic, zwitterionic and neutral species in the environment. PFCA and PFSA are often present as anionic forms, and they are water soluble and non-volatile, resulting in their environmental persistence. In contrast, neutral PFAS species (e.g., perfluoroalkyl sulfonamidoethanols (FASEs) and fluorotelomer alcohols (FTOHs)) are less water-soluble, more volatile and biodegradable than the ionic PFAS (Liu and Mejia Avendaño, 2013; Mejia Avendaño and Liu, 2015; Liu and Liu, 2016; Ochoa-Herrera et al., 2016). Because of their persistence, PFAS have been also detected in various biological samples (e.g., human serum and milk, cow milk, and fish samples) across geographical boundaries (Giesy and Kannan, 2002). PFCA and PFSA have been found ubiquitously in various environmental matrices including water (Ahrens and Bundschuh, 2014; Rahman et al., 2014; Banzhaf et al., 2017; Gebbink et al., 2017a), air (Langer et al., 2010; Goosey and Harrad, 2012; Jogsten et al., 2012), food (Haug et al., 2010; Eriksson et al., 2013; Domingo and Nadal, 2017), wildlife (Taniyasu et al., 2005; Kelly et al., 2009; Yeung et al., 2009; Houde et al., 2011; Galatius et al., 2013), and humans (Keller et al., 2010; Okada et al., 2013; Lee et al., 2018). Environmental monitoring studies have reported the presence of PFAS in plants (Scher et al., 2018; Zhang et al., 2018; Zhao et al., 2018), fishes (Chen et al., 2015), soils and minerals (Lee and Mabury, 2017).

To date, the global fate and transport pathways of ultra-short- and short-chain PFAS have not been well understood. Two main hypotheses have been proposed for their global transport: First,

that hydrophilic and ionic PFAS are transported directly by oceanic currents and/or by means of sea spray (Ahrens et al., 2010a; Cai et al., 2012a). Second, that neutral and volatile PFAS precursors undergo long-range atmospheric transport and are degraded/transformed in remote regions to ultra-short- and short-chain PFAS (Jahnke et al., 2007; Ahrens et al., 2011). Modeling studies mostly identify the ocean currents as the dominant transport pathway (Armitage et al., 2009). Meanwhile, measurements of ultra-short- and short-chain PFAS in the aqueous environment are still limited and further research is needed. Fig. 2 depicts the major pathways of PFAS in the environment. As summarized in Table 1, PFAS are frequently detected in all types of aqueous environments including seas, oceans (Yamashita et al., 2004, 2008; Cai et al., 2012b; Brumovský et al., 2018), rivers (Senthilkumar et al., 2007; Sharma et al., 2016; Gebbink et al., 2017b; Yeung et al., 2017a), surface/urban runoffs (Kim and Kannan, 2007; Awad et al., 2011; Meyer et al., 2011; Xiao et al., 2012b), drinking waters (Post et al., 2009; Thompson et al., 2011; Castiglioni et al., 2015; Hu et al., 2016), groundwaters (Moody and Field, 1999; Barzen-Hanson and Field, 2015; Baduel et al., 2017), rain/stormwater, deep polar seas, and snow (Taniyasu et al., 2008, 2013; Butt et al., 2010; Eschazier et al., 2010; Xie et al., 2015).

Currently, the most widely used short-chain PFAS are PFBS, perfluorobutanoic acid (C4, PFBA) and perfluorohexane sulfonic acid (C6, PFHxS) (Jensen and Warming, 2015). Studies have shown that these short-chain compounds and new generation replacement classes (e.g perfluoroethers) are equally persistent and bio-accumulative as their long-chain counterparts (Wilkinson et al., 2017a, 2017b; Gomis et al., 2018). For instance, researchers have recently employed X-ray photoelectron spectroscopy (XPS) to measure PFAS at the surface of consumer products and have found that used textiles were primarily composed of long-chain PFAAs, while new textiles were predominantly composed of short-chain PFAAs (Tokranov et al., 2018). Further, polyfluoroalkyl precursors (e.g. Fluorotelomer alcohols, Polyfluoroalkyl phosphoric esters and diesters, N-Alkyl perfluorooctane sulfonamides etc.) can go through environmental biotransformation/degradation into PFAS, partially contributing to their environmental concentration (Harding-Marjanovic et al., 2015; Mejia Avendaño and Liu, 2015; Ruan et al., 2015; Zhao et al., 2018). The limited information on the occurrence of short-chain PFAS in the environment could be attributed to limited data available about their precursors as well as the inherent difficulty in their identification (Zhou et al., 2013).

Despite their recent adoption, in some cases, the concentrations of short-chain PFAS in the environment were detected at much higher levels than the long chain compounds. For example, PFBS and PFBA in a Chinese Lake were found to be as high as 3660 ng/L and 4770 ng/L, respectively (Zhou et al., 2013), while in southern England water bodies that received sewage treatment effluents, concentrations of PFBS was observed to be 1.4–1.7-fold higher than the long-chain PFAS (Wilkinson et al., 2017b). Similar occurrences of elevated levels of short-chain PFAS have also been reported in sludge and solid matrices (Li et al., 2010; Yan et al., 2012). Studies of PFAS in ten wastewater treatment plants in the US found that the concentrations of 6:2 fluorotelomer sulfonate and perfluorooctanoic acid (PFOA) remained unchanged through trickling filtration, while those of PFOS and perfluorodecane sulfonic acid (PFDS) increased during activated sludge treatment (Schultz et al., 2006). Recently, modeling results of the global inventory of PFHxS (C6) have expected a continuous release of this short-chain PFAS by 1–21 ton between 2016 and 2030 (Boucher et al., 2017).

Similar to the occurrence of short-chain PFAS, the commercially employed fluorinated alternatives such as ADONA and GenX have also been detected in Cape Fear watershed, downstream of wastewater effluent discharges from manufacturing sites in NC,

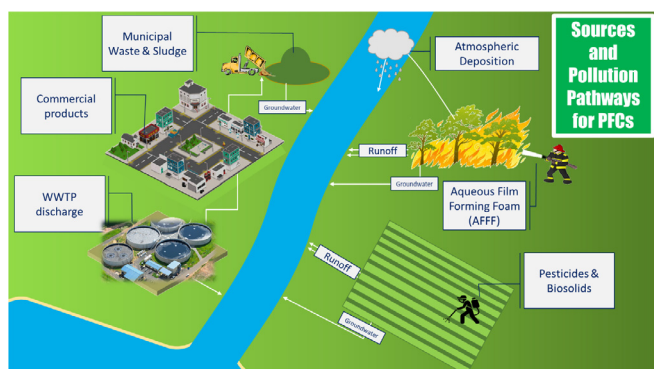


Fig. 2. Diagram of the sources and pollution pathways of PFAS in the environment.



USA (Wang et al., 2013b; Sun et al., 2016). In the Netherlands, the concentration of GenX in river and drinking water reached up to 812 ng/L (Gebbink et al., 2017a). Several sites along the Xiaoqing river in China were also found to have  $\Sigma$ GenX of ~9850 ng/L (Heydebreck et al., 2015a, b). The increasing trend of detection of known short-chain fluorinated alternatives is significant because according to the EPA's PFOA Stewardship program, there are over 150 potential short-chain and other alternates for PFOA 'alone' which cannot be feasibly detected with existing analytical technologies (Wang et al., 2017c). Therefore, future research should continue to focus on determining the occurrence of the ultra-short- and short-chain alternatives as well as the emerging perfluorinated classes in the environment.

The distribution of ultra-short- and short-chain PFAS and emerging fluorinated compounds in the global environment have been on the rise in recent years, due to their high solubility in water, low/moderate sorption to soils and sediments and resistance to biological and chemical degradation. Unfortunately, the lack of data prevents a clear understanding of the environmental mobility of these compounds. Precursors are likely to have different physical and chemical properties than their breakdown products, leading to differences in their transport behavior.

The previous studies on the toxicity PFAS considered only a few compounds. Thus, there is no agreement among the researchers on safe PFAS exposure levels yet (Kotthoff and Bücking, 2018). In their evaluation of the pharmacokinetics of PFBS in rats, monkeys, and humans, Olsen et al. (2009) noted a much lower potential for accumulation in human serum after repeated occupational, non-occupational (e.g., consumer), or environmental exposures (a single dose of 30 mg-PFBS/kg body weight). On the other hand, Oldham and coworkers reported that PFBS disrupted different model lipid assemblies indicating the potential of PFBS to be a human toxicant (Oldham et al., 2012). A fairly recent study reported that short-chain PFAS show a high global contamination potential due primarily to their environmental stability and mobility (Wang et al., 2015). Overall, additional studies are warranted to understand the environmental fate and potential impacts associated with the short- and ultra-short chain PFAS alternatives.

#### 4. Removal techniques from water: current practices and challenges

With the exception of a few studies entailing the examination of short-chain PFAS, most studies on PFAS removal from water have been limited to PFOS and PFOA (Table 3) Merino et al. (2016). In this article, we focus only on the studies involving the removal of ultra-short and short-chain PFAS.

##### 4.1. Adsorption by carbonaceous adsorbents

The adsorption on different carbonaceous materials (e.g., granular activated carbon (GAC), powdered activated carbon (PAC), and carbon nanotubes (CNT)) is the most studied technique for the removal of PFAS and other organic pollutants (Du et al., 2014; Shimizu et al., 2018). Hydrophobic and electrostatic interactions are the main two mechanisms for PFAS removal by adsorption on carbonaceous materials. As described previously, both short-chain and ultra-short chain PFAS are hydrophilic and less likely to form aggregates. For instance, the most commonly studied short-chain PFAS (i.e., PFBS, PFBA and PFHxA) have a  $\log K_{ow}$  (neutral form) = 2.82–4.6, water solubility > 20 g/L, and  $\log K_{oa}$  = 6–6.7,  $pK_a < 1$ , and  $\log K_{oc}$ : 2.7–3.6 (Alava et al., 2015). Further the decrease in  $\log K_{oc}$  values of PFAS with a corresponding decrease in the carbon chain length reduces the sorption potential of shorter chain PFAS (Higgins and Luthy, 2006; Ahrens et al., 2010b; Eschauzier

et al., 2012; Rahman et al., 2014). In all such previous studies, the effective removal of long-chain PFAS and lower removal efficiencies of short-chain PFAS was prevalent on GAC (Kothawala et al., 2017), activated carbon fibers (Chen et al., 2017), and CNT (Deng et al., 2012a). Further, the rate of removal of perfluorosulfonates acids (PFSA) was superior to that of the corresponding perfluorocarboxylic acids (PFCA) with the same chain length, as the former contains one more C–F bond resulting in stronger hydrophobic properties, and hence increased sorption (Zhou et al., 2010). Based on their physicochemical properties and relatively lower concentrations in natural waters, it can be hypothesized that the sorption of short-chain PFAS on adsorbents is more dependent on the availability of open active sites. Thus, the presence of natural organic matter (NOM) and other more hydrophobic compounds including long-chain PFAS and competitive ions in the water matrices will exhibit a greater effect on the rate of removal (Yu et al., 2012; Ateia et al., 2017a).

**Effect of NOM:** Very little data are available regarding the impact of NOM on the adsorption of short-chain PFAS by carbonaceous adsorbents. Indeed, the effect of NOM on adsorption of PFAS on carbonaceous materials and soil minerals is characterized by opposing observations. Specifically, the competitive occupation of NOM in both pore spaces and/or sorption sites have rendered them unavailable for sorption of PFAS. In independent bench-scale studies, Hansen et al. (2010) and Zhao et al. (2011) noted an adverse effect of NOM adversely upon the GAC adsorption of short- and long-chain PFAS (i.e.,  $C_4$ – $C_{10}$ ) due to both preloading and direct competition effects. In agreement with these observations, examination of full-scale plant data indicated frequent reactivation or replacement of GAC to maintain continuous removal of PFAS from water given the observed penetration of PFOS, PFHxS, PFBS, PFOA, PFHxA, and PFPA as early as 2–3 months into the operation (Hölzer et al., 2009). Similarly, co-existing organic compounds, specifically humic acid, 1-naphthol, phenol, and benzoic acid significantly affected the adsorption of PFOS on CNT (Deng et al., 2015). Of these, humic acid exhibited the most significant degree of suppression for PFAS adsorption on the MWCNT, followed by 1-naphthol, benzoic acid, and phenol. Regarding the chain length, the adsorbed amounts of different PFAS on the MWCNTs were a result of the increased order of PFBS ( $C_4$ ) < PFHxS ( $C_6$ ) < PFOA ( $C_8$ ) < PFOS ( $C_8$ ), consistent with their hydrophobicity (Deng et al., 2015). However, an increase in PFAS (i.e.,  $C_4$ – $C_{11}$ ) removal was recently observed with increase in NOM concentration in water (Kothawala et al., 2017). Although the authors did not elaborate on their observation, the formation of NOM–PFAS complexes is a possible explanation for this positive influence of NOM (Li et al., 2018).

**Effect of pH:** The existence of PFAS in a cationic, anionic and zwitterionic state supports the concept that the PFAS sorption behavior of PFAS is affected by pH, with changes in pH also influencing the electrostatic interactions between the sorbent and PFAS. However, as with NOM, little data are available regarding the effect of pH on the adsorption of short-chain PFAS compounds. The various conclusions of long-chain PFAS studies regarding the effect of pH have also complicated this concept. Specifically, although an increase in pH was found to increase the removal of PFAS (You et al., 2010; Zhi and Liu, 2016), no such relationship between the pH and adsorption affinity of PFAS was observed (Kwadijk et al., 2010; Milinovic et al., 2015; Barzen-Hanson et al., 2017a). Clearly, an expanded research program is needed to expand the findings of these limited endeavors.

**Regeneration of adsorbents:** Given that adsorbents used for PFAS removal are deemed hazardous, the regeneration and reuse of these spent adsorbents are most important. Methods to regenerate spent adsorbents, in general, include: 1) thermal and gas treatments, 2) chemical and extraction treatments, 3) biological

**Table 3**  
Summary of the currently available studies on removal of short-chain PFAS in the literature.

Category	Sorbent or Treatment Method	Target short-chain PFASs	Initial PFAS concentration (ng/L)	Water matrix	Reference	
Sorption	ACF SWCNT, MWCNT	PFBS	$10 \times 10^7$	Distilled Deionized Water (DDI) DDI	Chen et al. (2017) Deng et al. (2012b)	
		PFBA	$3-150 \times 10^6$			
	MWCNT GAC, PAC	PFBS	$75 \times 10^3$	0.25 mg/L Humic acid in DDI Groundwater (GW)	Deng et al. (2015) Hansen et al. (2010)	
		PFHxA				
		PFBS				
	GAC	PFHxA	$280 \pm 40$	GW	Zhao et al. (2011)	
		PFHpA	$320 \pm 40$			
	GAC	PFBA	$50 \times 10^6$	Surface water	Appleman et al. (2013)	
		PFBS				
	GAC	PFPeA	1000	DWTP	McCleaf et al. (2017)	
		PFHxA				
		PFBA				
		PFHxA				
		PFHpA				
	IX resin	PFBS	$1 \times 10^9$	DDI	Alessandro et al. (2016)	
PFBA						
PFBS		GW		Du et al. (2015)		
PFBA						
PFBS		$1.5 \times 10^8$		Industrial wastewater	McCleaf et al. (2017)	
PFHpA						
PFHxA						
PFBA						
Sorption	Non-ionic Resins Polymer (PEI-f-CMC)	PFBS	NA	DWTP, Italy Lake Water, USA	Contea et al. (2015) Ateia et al. (2018a)	
		PFBA	1000			
	Amine-f-COF	PFPeA	1000	DDI	Ji et al. (2018)	
		PFHxA				
		PFHpA				
		PFBS				
		PFHxS				
		PFBA				
	Membrane	NF	PFBS	1000	DDI and Artificial GW	Appleman et al. (2013)
			PFHxA			
	Oxidation/Mineralization	Electrochemical Mineralization	PFBS	$2.5-15 \times 10^4$	DDI	Wang et al. (2018a) Niu et al. (2012)
			PFBA	$10-15 \times 10^{10}$		
PFHxA						
Photolysis/Photocatalysis		PFHpA	$2 \times 10^9$	DDI	Zhuo et al. (2012)	
		PFPeA				
		PFBA				
Biological removal/ remediation	Plant uptake	PFBS	$12 \times 10^7$	DDI	Liao and Farrell (2009) Ravichandran et al. (2009)	
		PFBA	$4 \times 10^{11}$			
		PFHxA	$2-5 \times 10^{11}$	DDI	Ravichandran et al. (2007) (Hori et al., 2003, 2017) Felizeter et al. (2012a)	
		PFHpA				
		PFPeA				
		Growth medium solution.	PFBS	$200-2 \times 10^{12}$	Growth medium solution.	Müller et al. (2016)
			PFBA			
PFHxA						
PFHpA						
		PFPeA			Krippner et al. (2014)	

treatment, 4) vacuum treatment, 5) electric and electrochemical treatment, 6) supercritical fluid treatment, 7) hot water, 8) and microwave, ultrasounds,  $\gamma$ -ray irradiation and photochemical treatments (Deng et al., 2015; Ateia et al., 2018b; Wang et al., 2018a). Although thermal regeneration is the most commonly used technique by carbon manufacturers, it is ineffective in eliminating adsorbed PFAS from carbon surface given that C–F is the strongest single bond in organic chemistry after the B–F, Si–F and H–F single bonds (Atherton et al., 1996). Although the use of a higher regeneration temperature to break the C–F bond may remove PFAS from carbon surfaces, high losses of carbon often result (Sun et al., 2017). Thus, it is imperative to develop a series of regeneration methods that are more efficient than those available for the precise management of spent adsorbents after PFAS adsorption.

#### 4.2. Removal by other sorbents

Polymeric ion exchange (IX) resins consist of polystyrene or polyacrylic bead attached with a charged functional group balanced by a counter ion (cation/anion), a removal that occurs from electrostatic interactions leading to a counterion exchange with the charged PFAS entities. Conversely, non-ion exchange (NIX) resins are neutral synthetic polymeric structures that remove PFAS primarily by the non-ionic hydrophobic and van der Waals interactions. Although regulatory protocols detail the use of several types of resins in concentration and clean-up procedures during PFAS measurement (e.g. weak anion exchange resins), resins are rarely used for the removal of PFAS in water treatment facilities (Villagrasa et al., 2006; Shoemaker et al., 2008; Brumovský et al., 2018). Other newly developed sorbents that have been tested for the removal of short-chain PFAS include poly(ethylenimine)-functionalized cellulose microcrystals (PEI-*f*-CMC) (Ateia et al., 2018a) and amine-functionalized covalent organic frameworks (COF) (Ji et al., 2018).

Of the very few studies undertaken to examine the removal of short-chain PFAS, only a limited efficiency was observed regarding the removal of short-chain PFAS given their lower sorption potential, early breakthrough volumes and faster desorption (Higgins and Luthy, 2006; Chularueangaksorn et al., 2013, 2014; Lundgren, 2014). It is also possible that long-chain PFAS may also outcompete shorter-chain analogs when they coexist in sorption processes, due to the addition of extra  $\text{CF}_2$  moieties that increase the hydrophobicity of PFAS (Deng et al., 2012a; Du et al., 2015). Clearly, these findings indicate the challenge of removing short-chain PFAS (notably PFBA and PFBS) as compared to their long-chain counterparts.

The efficacy of IX resins used for the removal of short-chain PFAS has been hypothesized as dependent upon the charge, pore size and hydrophobicity of the polymer material (Alessandro et al., 2016; Schuricht et al., 2017). In their comparison of the sorption kinetics of PFBS ( $\text{C}_4$ ) and PFOS ( $\text{C}_8$ ) on two polystyrene gel resins, Deng et al. (2010) noted a higher removal of PFBS than PFOS. In another study investigating the efficacy of a strong basic anionic polystyrene-DVB cross-linked resin, McCleaf et al. (2017) reported >80% removal of short-chain PFAS ( $\text{C}_4$ – $\text{C}_7$ ) with an initial concentration of 2.5  $\mu\text{g/L}$ . However, when the same resin was used to remove PFAS at lower concentration (<100 ng/L), desorption of the compounds was observed at early breakthrough volumes. Similar trends in short-chain PFAS ( $\text{C}_4$ – $\text{C}_7$ ) desorption were observed in pilot-scale experiments using groundwater (Alessandro et al., 2016). The same observations were reported for PEI-*f*-CMC (Ateia et al., 2018a) and amine-functionalized covalent organic frameworks (Ji et al., 2018).

**Competing Ions:** The presence of anions (e.g., sulfate, chromate,

nitrate, chloride, and perchlorate) in natural waters with concentrations at orders of magnitude higher than PFAS results in a significant competition with PFAS for the exchange sites (Alessandro et al., 2016). Further, desorption of the short-chain PFAS may result from the displacement by NOM or other competing molecules including long-chain PFAS (Higgins and Luthy, 2006; Eschauzier et al., 2012). However, Dudley (2012) reported an increase in the removal of PFAS ( $\text{C}_3$ – $\text{C}_9$  PFCA and  $\text{C}_4$ – $\text{C}_8$  PFSA) by IX resins in the presence of NOM. They established two hypotheses for this phenomena: 1) that the PFAS associated with NOM were pulled into the resin matrix, and 2) that NOM modified the resin surface in such a way that increased the affinity for PFAS. Despite these novel findings, our understanding of the effect on NOM on short-chain PFAS removal by resin is still limited, which necessitates a comprehensive and the role of NOM type and concentration (Ateia et al., 2017b).

**Effect of pH:** Although the effect of pH on the exchange capacity of resins has not been investigated extensively, it has been observed that PFAS sorption via resin does typically increase with a decrease in pH (Higgins and Luthy, 2006; McKenzie et al., 2015).

**Regeneration:** Commercially available IX resins are often regenerated using liquid rinses composed of either methanol or brine solutions (Du et al., 2014). Further, as NIXs usually produce weaker substrate adsorptive binding, resin regeneration is often less intensive than those required for IXs (Senevirathna et al., 2010). However, given that brine solutions are byproducts from the regeneration of IX resins with high PFAS concentrations, (Senevirathna et al., 2011; Xiao et al., 2012a; Chularueangaksorn et al., 2013). Further, Although single-use IX resins are now available for PFAS removal (Woodard et al., 2017), their application is expensive. Therefore, novel destruction methods for managing IX regeneration solutions for both short- and long-chain PFAS are needed to make this technology more applicable.

#### 4.3. Other treatment technologies

**Removal by membranes:** High-pressure membrane processes such as reverse osmosis (RO) and Nanofiltration (NF) membranes are most effective for removing various classes of trace organic compounds from water (Fujioka et al., 2014). However, very little data are available regarding the use of membranes in the removal of short-chain PFAS (Appleman et al., 2013; Wang et al., 2018a). Generally, the rejection rate of NF membranes is high (>95%) for molecules with molecular weight more than 150 Da (Eriksson, 1988), which is a suitable range for all short-chain PFAS. In one study undertaken to examine short-chain PFAS removal, a reduction in pH was observed to increase the membrane rejection of short-chain PFAS (Zeng et al., 2017), inferring the importance of pH plays in the rejection of PFAS. Ionic strength can also alter the extent of the Donnan-exclusion, which is thus involved in altering the membrane rejection characteristics for negatively charged PFAS. In addition, the organic membrane fouling builds up the cake layer on the membrane surface, which again influences the PFAS rejection. Similar to ion exchange processes, membrane technologies are not destructive and generate a high PFAS concentrated brine solution, which requires treatment or disposal.

**Advanced oxidation processes (AOPs):** Oxidation methods have been used to decompose PFAS with varying success rates. Most studies have reported treatment results with either PFOA or PFOS as the representative compound. Among the various tested AOPs for PFAS removal (chemical oxidation, photolysis, photocatalysis, sonochemical degradation), electrochemical oxidation has the advantages of strong oxidation and low energy consumption with higher removal efficiency for short-chain PFAS (Niu et al., 2012). The most commonly investigated electrodes for PFAS

removal include boron-doped diamond (BDD) thin film, Ti/SnO<sub>2</sub>, Ce/PbO<sub>2</sub>, and Ti/RuO<sub>2</sub>. Electrochemical oxidation of short-chain PFAS in aqueous solution on a BDD anode showed that decomposition of PFBA, PFHxA, PFBS, PFHxS occurred with direct transfer of one electron from the carboxyl or sulfonate group to BDD forming PFAS radicals resulting in a defluorination reaction between perfluoroalkyl radical and hydroxyl radical (Zhuo et al., 2012). Using a low current density and high PFBS concentration (>150 mg/L), (Liao and Farrell, 2009) were able to achieve >90% removal efficiency with Si/BDD electrode within an hour. (Niu et al., 2012) reported that the efficacy of electrochemical oxidation of PFAS drops with the decrease of chain-length (e.g., the removal efficiency decreased from 98% for PFHpA to 32% for PFBA after 1.5 h at an initial concentration of 100 mg/L). This decrease in the removal efficiency of short-chain PFAS could be attributed to the fact that electrochemical degradation of the co-existed long-chain PFAS led to the formation of shorter-chain PFAS (Trautmann et al., 2015). The generation of •OH radical depends on electrode potential, current density, pH, plate distance, electrolyte, and the initial concentration are the most influencing factor on electrochemical oxidation techniques. These factors also directly contribute to the service life span of electrodes and the mass transfer rate of PFAS on to the electrodes.

Photocatalytic degradation is another AOP that refers to the use of ultraviolet light with a photocatalyst to decompose/mineralize target compounds. Hori et al., (2003) and Hori et al., (2017) reported on the degradation of trifluoroacetic acid (TFA) using photocatalytic activity, however, the initial TFA concentration was 18–20 × 10<sup>9</sup> ng/L. Yet, no available systematic studies on the photocatalytic degradation of ultra-short chain and short-chain PFAS at environmentally relevant conditions. Although there are several reports indicating the degradation of PFOA and fluorotelomer compounds via a radical based oxidative/reductive mechanisms (Sahu et al., 2018), PFOS appear to be significantly more resistant to chemical oxidation. Lastly, It must be highlighted that AOPs always result in the generation of significant concentrations of more hydrophilic short-chain and ultra-short chain PFAS from long-chain precursors (Wang et al., 2017a).

**Biological remediation:** Previous studies that monitored wastewater treatment plants have frequently reported on elevated concentrations of short-chain PFAS due to the biotransformation of short-chain PFAS precursors in aerobic treatment or in membrane bio-reactors (Lee et al., 2010b; Pan et al., 2016). To date, however, there is no documentation of microbial degradation of short-chain PFAS. Meanwhile, as discussed in the above sections, PFAS tend to bioaccumulate in aquatic and terrestrial organisms. Therefore, PFAS can be removed from water by the means of plant uptake. Recent studies have shown that both the short-chain and long-chain PFAS tend to bioaccumulate in plant tissues (Felizeter et al., 2012b). Surprisingly, PFAS bioaccumulation was observed to follow a U shaped trend such that the highest accumulation occurs for the least sorptive (short-chain such as PFBA and PFPeA) and the most sorptive (longest-chain such as PFNA and PFDA) PFAS (Müller et al., 2016). Interestingly, of the PFAS studied, PFBA (C<sub>4</sub>) had the highest mean uptake rate. The higher accumulation of PFBA (C<sub>4</sub>) and PFPeA (C<sub>7</sub>) in plants has been attributed to small molecular size, as they may be co-absorbed passively with water. Another study that investigated the uptake and distribution of PFAS in maize (*Zea mays*) has concluded that the plant uptake and distribution of PFASs are dependent on chain length, functional group and plant tissue (Krippner et al., 2014). Accordingly, short-chain PFAS translocated predominantly to the shoots and the long-chain PFAS transferred predominantly to the roots by different plant species. Meanwhile, it was also suggested the uptake rate is both time- and concentration-dependent. Interestingly, a higher dose of PFAS

(1 mg/L) resulted in a lower percentage of plant uptake indicating PFAS toxicity (Garcia-Valcarcel et al., 2014).

## 5. Conclusions and future research directions

Although the increased use of ultra-short chain and short-chain PFAS has generated concerns regarding these compounds, little data exist concerning their transport, fate, and options for treatment. The high solubility of short-chain PFAS in water, low/moderate sorption to soils and sediments and resistance to biological and chemical degradation has resulted in their widespread presence in various aquatic environments. Further, improvements in analytical instruments and determination methods have made it possible to measure previously undetected PFAS. However, with the exception of a handful of studies examining the removal of several discrete short-chain PFAS from water, most studies are been limited to PFOS and PFOA. Consequently, future studies must be undertaken to expand upon the following topics:

- Comprehensive surveys of the occurrence of short-chain PFAS are needed, given the relative age of available data with the exception of some recent studies on specific contaminated locations.
- The occurrence data should be integrated to develop models to estimate of mass balance of PFAS in different matrices (e.g., land, water, air) and in sub-divisions within water environment (e.g., sea/oceans, rivers, lakes, snow, etc.).
- Researchers should consider elucidating the exposure and toxicity effect of short-chain PFAS on humans and other organisms. In addition, more efforts should be directed to finding the pathway for the migration of short-chain PFAS from commodities into drinking water.
- Although there are more than 3000 commercially available PFAS, studies are limited to small numbers of compounds. Thus, more efforts should be directed towards the identification and characterization of those unknown compounds.
- Selective, cheap, and scalable treatment alternatives must be developed as countermeasures to increasingly larger concentrations of short-chain PFAS in water environments.
- Studies must be undertaken to develop treatment options for the decontamination of short-chain PFAS under environmentally relevant concentration levels and conditions.
- Although sorption techniques are the protocols most extensively used, the competition from NOM and long-chain PFAS can negatively influence the removal of short-chain PFAS. Therefore elucidating the nature and impact of these competitions warrant further investigations. Hybrid sorption systems of (GAC + IX) may provide some viable alternatives to manage such competitions and enhance the removal of short-chain PFAS from the water. The effect background matrix (e.g., pH, competing anions) on the removal of PFAS by sorption processes also deserve further investigations.
- Although destruction methods (e.g. AOPs, advanced reduction processes (ARPs), electrochemical oxidation, etc.) are promising, they are expensive and difficult to scale, thus restricting their use to industrial applications where short-chain PFAS occur in high concentrations. Therefore, new catalysts that can remove low concentrations of short-chain PFAS in the presence of NOM and other coexisting ions must be developed and validated.
- Recent studies showed amine-functionalized materials as promising sorbents for different classes of PFAS. Yet, more research is needed on the development of new stable polymers with low desorption of short-chain PFAS.



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