

Recent developments in polyfluoroalkyl compounds research: a focus on human/environmental health impact, suggested substitutes and removal strategies

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Abstract Between the late 1940s and early 1950s, humans manufactured polyfluoroalkyl compounds (PFCs) using electrochemical fluorination and telomerisation technologies, whereby hydrogen atoms are substituted by fluorine atoms, thus conferring unnatural and unique physicochemical properties to these compounds. Presently, there are wide ranges of PFCs, and owing to their bioaccumulative properties, they have been detected in various environmental matrices and in human sera. It has thus been suggested that they are hazardous. Hence, this review aims at highlighting the recent development in PFC research, with a particular focus on perfluorooctanoate (PFOA) and perfluorooctane sulfonate (PFOS), the most studied and predominantly found PFCs in various environmental matrices, although recent reports have included perfluorobutane sulfonate (PFBS), which was previously regarded as innocuously harmless, when compared to its counterparts, PFOA and PFOS. As such, proper investigations are thus required for a better

understanding of short-chain PFC substitutes, which have been suggested as suitable replacements to long-chained PFCs, although these substitutes have also been suggested to pose various health risks comparable to those associated with long-chain PFCs. Similarly, several novel technologies, such as PFC reduction using zero-valent iron, including removal at point of use, adsorption and coagulation, have been proposed. However, regardless of how efficient removers some of these techniques have proven to be, short-chain PFCs remain a challenge to overcome for scientists, in this regard.

Keywords Polyfluoroalkyl compounds · PFOA · PFOS · PFBS · Substitutes

Introduction

Polyfluoroalkyl compounds (PFCs) are a wide assortment of anthropogenic chemicals, manufactured between the late 1940s and early 1950s (Niu et al. 2016) using electrochemical fluorination and telomerisation (Benskin et al. 2012; Banks et al. 2013). Thus, $F(CF_2)_xR$ is regarded as the general molecular formula for these chemicals, with two distinctive subsets characterising them, namely, PFCs, in which the head group contains no C-H bonds, and fluorotelomers (FT) in which the R-group contains an even-numbered alkyl-chains, resulting in the general formula of $F(CF_2)_x(CH_2-CH_2)_yR$ and $F(CF_2)_x(CH=CH)_yR$ (Møskeland 2010). Table 1 provides a general illustration of PFCs that have been of interest.

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Table 1 PFCs of interest, including their chemical structures and general formula (Butt et al. 2014; Kwon et al. 2016; Zhou et al. 2016)

Class	Compound	Abbreviation	General formula	
Polyfluorinated sulfonamides (FSAs)	<i>N</i> -methyl perfluorobutane sulfonamidoethanol	NMeFBSE	$F(CF_2)_4SO_2N(CH_3)CH_2CH_2OH$	
	<i>N</i> -ethyl perfluorobutane sulfonamidoethanol	NEtFBSE	$F(CF_2)_4SO_2N(CH_2CH_3)CH_2CH_2OH$	
	Perfluorooctane sulfonamide	PFOSA	$F(CF_2)_8SO_2NH_2$	
	<i>N</i> -methyl perfluorooctane sulfonamide	NMeFOSA	$F(CF_2)_8SO_2N(CH_3)H$	
	<i>N</i> -ethyl perfluorooctane sulfonamide	NEtFOSA	$F(CF_2)_8SO_2N(CH_2CH_3)H$	
	<i>N</i> -methyl perfluorooctane sulfonamidoethanol	NMeFOSE	$F(CF_2)_8SO_2N(CH_3)CH_2CH_2OH$	
	<i>N</i> -ethyl perfluorooctane sulfonamidoethanol	NEtFOSE	$F(CF_2)_8SO_2N(CH_2CH_3)CH_2CH_2OH$	
Fluorotelomer alcohols (FTOHs)	4:2 fluorotelomer alcohol	4:2 FTOH	$F(CF_2)_4CH_2CH_2OH$	
	6:2 fluorotelomer alcohol	6:2 FTOH	$F(CF_2)_6CH_2CH_2OH$	
	8:2 fluorotelomer alcohol	8:2 FTOH	$F(CF_2)_8CH_2CH_2OH$	
	10:2 fluorotelomer alcohol	10:2 FTOH	$F(CF_2)_{10}CH_2CH_2OH$	
	12:2 fluorotelomer alcohol	12:2 FTOH	$F(CF_2)_{12}CH_2CH_2OH$	
Perfluorosulfonates (PFSAs)	Perfluorobutane sulfonate	PFBS	$F(CF_2)_4SO_3^-$	
	Perfluorohexane sulfonate	PFHxS	$F(CF_2)_6SO_3^-$	
	Perfluorooctane sulfonate	PFOS	$F(CF_2)_8SO_3^-$	
	Perfluorodecane sulfonate	PFDS	$F(CF_2)_{10}SO_3^-$	
Perfluorocarboxylates (PFCAs)	Perfluorohexanoate	PFHxA	$F(CF_2)_5CO_2^-$	
	Perfluoroheptanoate	PFHpA	$F(CF_2)_6CO_2^-$	
	Perfluorooctanoate	PFOA	$F(CF_2)_7CO_2^-$	
	Perfluorononanoate	PFNA	$F(CF_2)_8CO_2^-$	
	Perfluorodecanoate	PFDA	$F(CF_2)_9CO_2^-$	
	Perfluoroundecanoate	PFUA	$F(CF_2)_{10}CO_2^-$	
	Perfluorododecanoate	PFDoA	$F(CF_2)_{11}CO_2^-$	
	Perfluorotridecanoate	PFTriA	$F(CF_2)_{12}CO_2^-$	
	Perfluorotetradecanoate	PFTetA	$F(CF_2)_{13}CO_2^-$	
	Perfluoropentadecanoate	PFPA	$F(CF_2)_{14}CO_2^-$	
	Perfluorohexadecanoate	PFHxDA	$F(CF_2)_{15}CO_2^-$	
	Fluorotelomer carboxylates (FTCAs, FTUCAs)	6:2 fluorotelomer carboxylate	6:2 FTCA	$F(CF_2)_6CH_2CO_2^-$
		6:2 fluorotelomer unsaturated carboxylate	6:2 FTUCA	$F(CF_2)_6CHCO_2^-$
8:2 fluorotelomer carboxylate		8:2 FTCA	$F(CF_2)_8CH_2CO_2^-$	
8:2 fluorotelomer unsaturated carboxylate		8:2 FTUCA	$F(CF_2)_8CHCO_2^-$	
10:2 fluorotelomer carboxylate		10:2 FTCA	$F(CF_2)_{10}CH_2CO_2^-$	
10:2 fluorotelomer unsaturated carboxylate	10:2 FTUCA	$F(CF_2)_{10}CHCO_2^-$		
Fluorotelomer sulfonates (FTSs)	6:2 fluorotelomer sulfonate	6:2 FTS THPFOS	$F(CF_2)_6CH_2CH_2SO_3^-$	
	8:2 fluorotelomer sulfonate	8:2 FTS	$F(CF_2)_8CH_2CH_2SO_3^-$	
	10:2 fluorotelomer sulfonate	10:2 FTS	$F(CF_2)_{10}CH_2CH_2SO_3^-$	

Moreover, there are various PFCs, of which two types have been the widely utilised by a variety of industries.

These are perfluorocarboxylic acids (PFCAs), identifiable by their structures, $F(CF_2)_xCOOH$, and perfluorosulfonic

acids (PFSA), $F(CF_2)_xS(O_3)H$. These PFCAs and PFSAs are acids which are readily ionised, thus can be negatively charged due to the loss of a proton, leading to their being referred to as perfluorocarboxylates and perfluorosulfonates, respectively (Schröter-Kermani et al. 2013). The most researched and reported, particularly in ecotoxicology studies, are perfluorooctanoic acid (PFOA, $F(CF_2)_7COOH$) and perfluorooctosulfonic acids (PFOS, $F(CF_2)_8S(O_3)H$) (Mudumbi et al. 2014a, b, c; Zhao et al. 2015; Shoeib et al. 2016; Yang et al. 2016a, b). Recently, perfluorobutane sulfonate (PFBS, $C_4HF_9O_3S$) has also been suggested to be a persistent organic pollutant (POP) once it enters the environment (Zhao et al. 2015; Shoeib et al. 2016; van den Dungen et al. 2016). In production techniques for these fluorocarbons, the substitution of hydrogen atoms by fluorine atoms from suitable precursors allows for the conferring of particular physicochemical properties to the compounds (Hidalgo and Mora-Diez 2016), such as chemical stability, non-wetting, fire, including weather resistance, and hydrophobicity and oleophobicity. They can lower the surface tension of viscous matrices, are irradiation resistant and biologically non-biodegradable (Ludwicki et al. 2015; Bennett et al. 2015; Niu et al. 2016), thus their persistency in the environment.

Molecular structure of polyfluoroalkyl compounds

Polyfluoroalkyl compound molecules are characterised by a perfluorinated carbon chain coupled with one special functional group at the end of the molecular chain, which can be either a carboxylic ($COOH$) or sulfonic group. The fluorinated carbon chain of PFCs directly influences their hydrophobicity, while the functional group permits the molecules to be hydrophilic. Different functional groups have shown diversified behaviour once introduced into different environments (Senevirathna 2010). Thus, some PFCs, that is, predominantly PFOA and PFOS, have been detected as residues in various environmental matrices, although current research has abundantly indicated that other PFCs, such as PFBS, should not be ignored.

Diversified application of polyfluoroalkyl compounds

Since PFCs have been manufactured for various applications due to their unique physical properties (Hagenaars et al. 2011), to date, numerous industries

have used these molecules as building blocks to form fluorinated polymers such as perfluoroalkylpolymers. These polymers should not be confused with fluoropolymers, such as polytetrafluoroethylene, that is, Teflon™, which are aliphatic compounds (Møskeland 2010; Ebnesajjad 2013). In certain cases, PFCs are used in the manufacturing process of fluoropolymers and later appear as residues in the final product (Herzke et al. 2007; Møskeland 2010). This, in our opinion, has diversified their utilisation, which exacerbates their prevalence, even in areas presumed free of such contaminants.

PFC-generated fluoropolymers are used as additives in hydraulic fluids, photographic emulsifiers and paints, to lower their surface tension, and/or as coating in carpets and textiles to allow stain and water repellency (Herzke et al. 2007; Møskeland 2010; Martens 2013). Furthermore, an exceptional and important application of PFCs has been in specialised aqueous film-forming foams (AFFFs) due to their ability to form films even at high temperature, a requirement when extinguishing fires (Place and Field 2012; Sha et al. 2015). Due to their versatility, various other industrial applications and processes have since been developed, thus giving rise to new products such as lubricants and motor oil additives, sports clothing, medical equipment, extreme weather military uniforms and waterproof breathable fabrics (Bao et al. 2014; Wang et al. 2014a, b; Niu et al. 2016). PFCs have also been used as polymerisation aids in the production of components for electronic products (Senevirathna 2010). Therefore, such diversified applications of these materials can result in far-reaching consequences, including consistent and prolific release, as well as transportation into living organisms. Tables S1 and S2 highlight various polymers and non-polymers which have been extensively used in several industry applications worldwide (provided as supplementary material, together with Tables S3–S6).

Polyfluoroalkyl compounds in the environment: discharge, transportation, occurrence and persistence

Discharge of PFCs directly into the environment

Consequently, as a result of excessive use, PFCs have found ways into the environment. As such, it has been reported that PFCs are discharged into the natural

environment both directly and indirectly (Wang et al. 2014a, b, 2015a, b). Thus, direct discharge has been regarded as the primary mechanism by which PFCs enter the environment from their life cycle (that is, manufacture, usage and disposal) when assessing their products, derivatives, residues or as unintentional by-products, that is impurities in consumer products (Li et al. 2015; Kotthoff et al. 2015). Their indirect discharge is suggested to be through transformation and/or degradation resulting in their presence in wildlife and humans (Guzmán et al. 2016; Gomis et al. 2016), as well as from fluorotelomer-made products through abiotic or biotic processes (Butt et al. 2014).

It has further been indicated that PFCs and their by-products including precursors may enter the environment via various other routes, such as (a) spilled discharge or through solid waste, for example exhaust/fuel gases from combustion, domestic wastewater, sludge and from manufacturing premises (Li et al. 2015; Kwon et al. 2016; Bečanová et al. 2016); (b) either by volatilisation along the supply chain from manufacturers to downstream industrial or end-consumers (OECD 2013; Oliaei et al. 2013); (c) or through fugitive release by end-users, especially where PFC-containing products (for example, fluoropolymer manufacturing sites, paper and textile factories) including their precursors have been processed into final products (Kotthoff et al. 2015). Furthermore, their incorporation into raw materials/consumer products can result in their wash-off directly into the environment (Kotthoff et al. 2015; Bečanová et al. 2016). In most cases, unsuitable treatment methods are applied. For instance, the use of sewage sludge as a fertiliser, untreated outgassing from landfills or insufficient wastewater treatment, can further exacerbate contamination of PFC-free environments or the food chain (Gallen et al. 2016; Kwon et al. 2016).

Occurrence, transportation and persistence of polyfluoroalkyl compounds

Polyfluoroalkyl compounds, especially PFOA and PFOS, have been known to display both persistence and long-range transportation (LRT) once they have entered the environment. This has been confirmed by their ubiquitous presence in various environmental matrices far away from anthropogenic activities (Stock et al. 2010). However, the fact that PFCs have different properties than their counterparts, that is BFRs and

PCBs for which models of environmental persistence and LRT have been developed, result in the complexity of developing suitable models for their persistence and LRT, which can conclusively explain the mechanisms of how PFCs are transported in the environment (Møskeland 2010). This is because PFCs (that is PFOA and PFOS) are strong ionic acids and surface wetting agents, as opposed to being hydrophobic apolar compounds, characteristics associated with BFRs and PCBs (Fliedner et al. 2012). The pK_a (or acid-dissociation constant) of these substances has been estimated to be near 0 for PFCAs, for example PFOA, and around -3 for PFSAs, for example PFOS (Campbell et al. 2009; Møskeland 2010), making them some of the most effective surfactants.

Additionally, it has been indicated that the perfluoroalkyl tail of these substances is one of the most hydrophobic molecular fragments and anionic/acidic functional groups (CO_2^- , SO_3^-). Consequently, it has been suggested that PFCAs and PFSAs have a strong affinity to water with a hydrophilic head, whereas the rest of the molecule is hydrophilic (Xiao et al. 2013). Thus, these molecules are likely to have a high LRT in the environment through water transportation (for example, by dispersion in lakes and rivers, including sorption to atmospheric moisture) as previously indicated by some studies (Schindler et al. 2013; Shan et al. 2015; Kirchgorg et al. 2016).

According to Yao et al. (2015) and Guo et al. (2015), the uniqueness of PFCs and the mechanism of their transportation into the environment have remained an active area of research, and for this reason, part of the recommendations proposed includes scientists being able to deal with the unique environmental transportation and partitioning processes of PFCs, that is, that researchers need an additional set of model parameters to account for the ionic and surfactant nature of these compounds, their pK_a (the acid-dissociation constant) and surface-water sorption coefficients, including their critical micelle and aggregate-formation concentrations (Zhou et al. 2010a; Zareitalabad et al. 2013).

Recent research has since reported the distribution of PFCs globally (Rankin et al. 2015; Washington and Jenkins 2015a; Routti et al. 2015). Overall, PFCs have been found in surface river waters, or alternatively, in wastewater treatment plants in South Africa (Mudumbi et al. 2014b; Adeleye 2016; Chen et al. 2016; Pitarch et al. 2016; Shiwaku et al. 2016; Lopez et al. 2015; Lescord et al. 2015; Lu et al. 2016; Hu et al. 2016;

Zhang et al. 2016). It also has been indicated that water currents and evaporation/precipitation have facilitated the transportation of these substances into remote areas, such as the arctic, remote islands and other remote inland environments, for example alpine lakes, etc. (Lescord et al. 2015; Wang et al. 2015a, b; Yamazaki et al. 2016). Additionally, evidence suggests that among all environmental media, the ocean is likely to be the largest global reservoir of PFCs such as PFOA (Cousins et al. 2011); thus, inland deposition through the water cycle is inevitable.

PFOA and PFOS have dominated most reports, with PFOS being found in higher concentration levels, that is, 271.10 g/L, in a recent study from Spain (Campo et al. 2015). Moreover, PFBS has recently received attention among the list of PFCs with researchers believing that it should not be overlooked, with Zhou et al. (2013) indicating that although PFBS have a lower adsorption potential than PFOA and PFOS, which suggests its lower potential to bioaccumulate in aquatic biota, its aquatic and ecological risk must be assessed, because of the substance's increasing usage, release and transportation.

Over the past two decades, research projects have demonstrated the susceptibility of living organisms to PFCs. They have thus been detected in human sera (Ludwicki et al. 2015; Shrestha et al. 2015), animals (Filipovic et al. 2015; Koponen et al. 2015) and plants (Mudumbi et al. 2014c; Blaine et al. 2014a, b; D'Hollander et al. 2015; Yang et al. 2015). Additionally, some reports have indicated that, although fluorotelomer alcohols (FTOHs), fluorotelomer sulfonamides (FSAs) and fluorotelomer sulfonic acids (FTS) have been regarded as the most substantial PFC precursors, several hundred other PFCs are considered to be capable of conversion into PFCAs and PFSA (Gomis et al. 2015; Sun et al. 2016). Additionally, precursors to PFCs, such as FTOHs, are volatile and can be released from products under ambient conditions and later be transformed into PFCs (EPA 2014). As a result, it has been argued that the occurrence of PFCs and its salts is not only due to direct release of these compounds into the environment but is also due to the indirect conversion of many other PFCs (Kim et al. 2015). It has also been indicated that both direct and indirect sources of these compounds were considered in multimedia models that account for the occurrence of these substances (Kim et al. 2015; Gomis et al. 2015), with the modelling of PFOA distribution and its higher

homologues being reported in a review (Cousins et al. 2011). The models were generally found to support the conclusion that direct use of PFOA and PFOS-based products was the dominant global environmental contributor for these two PFCAs (OECD 2013).

Polyfluoroalkyl compounds' precursors of concern

Various reports have suggested that PFCs enter the environment by either direct or indirect sources. Hence, direct sources are regarded as the discharge of PFCs into the environment as such, regardless of whether it is intentional release or otherwise (Buck et al. 2011; Liu 2015), while indirect sources imply the formation of PFCs by means of biotic or abiotic degradation from other perfluoroalkyl and polyfluoroalkyl substances (PFASs), regarded, in this case, as precursors to PFC (pre-PFCs), as they enter various environmental mediums (Buck et al. 2011; Liu 2015). Thus, researchers believe the indirect sources play a significant role in the prevalence of PFCs in humans and the environment (Benskin et al. 2013; Lee et al. 2014; Liu 2015; Avendaño and Liu 2015). The OECD released a list of 615 pre-PFCs which have the potential to degrade into PFCA (OECD 2007). Table S3 depicts examples of these types of substances, most of which there is limited data available on their pathways into the environment.

Furthermore, examples of pre-PFCs have included mono- and di-esters such as sodium 2-(N-ethylperfluorooctane-1-sulfonamide) ethyl phosphate (SAmPAP), sodium bis-[2-(N-ethylperfluorooctane-1-sulfonamido) ethyl] phosphate (diSAmPAP), N-ethyl perfluorooctane sulfonamide (EtFOSA), etc. According to Wellington Report (2014), not only are SAmPAP esters persistent in the ecosystem, but they are precursors of PFOS, and very little evidence is available on their lifetime and transformation. Hence, it has been indicated that most PFAS-containing products that humans rely on daily are rather containing pre-PFCs (Herzke et al. 2012; Gebbink et al. 2013; Liu 2015), and which, according to available data, have not been investigated (Wellington Report 2014; Liu 2015), suggesting these precursors being potential threats to the consumers. For instance, the PFOS-precursor EtFOSA is used in the manufacturing of sulfluramid, a pesticide for controlling leaf-cutting ants (Löfstedt Gilljam et al. 2015). Ultimately, this explains why PFOS has largely been detected in the environment, with its plant concentration levels higher in certain countries, like in South

Africa (Mudumbi et al. 2014b), where agriculture is an integral part of the economy. Similarly, a lengthy biodegradation half-life of *N*-ethyl perfluorooctane sulfonamido ethanol (EtFOSE), another pre-PFOS, and the recalcitrant nature of SAMPAP were recently reported by Benskin et al. (2013), and which, according to the authors, explains the elevated concentrations of PFOS-precursors in the environment. However, it is argued that clarity is needed on whether SAMPAP can be a potential significant source of PFOS in benthic and higher trophic level organisms (Benskin et al. 2013). It has been further suggested that the development of enhanced (i.e., residual-free) SAMPAP standards would be of great assistance to scientists who assess the stability and environmental behaviour of these substances (Benskin et al. 2013).

On the other hand, recent data has revealed the potential of fluorotelomer-based polymers to degrade and to form PFOA and related compounds (Washington et al. 2015b). Hence, researchers have suggested that elevated concentrations of pre-PFCs observed in studied samples explain the large distribution of PFCs in the natural environment and beyond, i.e. to areas far from their production (Benskin et al. 2013; Washington et al. 2015b), and these precursors thus might constitute the major sources of PFOA, PFOS, etc. (Washington and Jenkins 2015a) but have also called for more investigations to be conducted (Washington et al. 2015b).

Bioaccumulation of PFCs in biota and humans

Bioaccumulation potentials are estimated using what is known as the partition coefficient (K_{ow}) between octane-water phases (OECD 2013). However, because PFCs are surfactants, an emulsion can be formed during measurements. It has been reported that K_{ow} is unknown for most PFCs (OECD 2013). Therefore, to determine the bioaccumulation potential of PFCs in environmental media, either a bioaccumulation factor (BAF) or a bioconcentration factor (BCF), which is the extent to which pollutants concentrate from water into other matrices (Chiou 2003), can be estimated by dividing the average concentrations in matrices by the concentrations of PFCs in a water environment (Senevirathna 2010). BAF or BCF should not be confused with biomagnification factor (BMF) used to refer to the ratio of contaminant concentration in biota to that in the surrounding water when the biota was exposed via contaminated food (Nowell et al. 1999). It is determined

by dividing the average concentrations in predators to those in prey (Senevirathna 2010).

As a result, BMF has been quantified globally in various species, particularly in fish (Lescord et al. 2015; Ahrens et al. 2015; Hong et al. 2015; Bossi et al. 2015; Svihlikova et al. 2015, Ahrens et al. 2016), polar bears (Letcher et al. 2014; Jenssen et al. 2015), including albatross (Chu et al. 2015), and seals (Routti et al. 2015), to name a few, with results indicating that long-chained PFCs are bioaccumulative (Kakuschke and Griesel 2016; Zhai et al. 2016) and can ultimately biomagnify in the food chain (Zhang et al. 2015; Franklin 2016) and in humans (Fujii et al. 2015; Goudarzi et al. 2016). Table S4 reports on the bioaccumulation potential (BMF) of selected PFCs in certain aquatic organisms.

As such, various PFASs and PFCAs have been detected in human sera in the general population (Bennett et al. 2015; Gomis et al. 2016) of which PFOA, PFOS and PFBS are the most frequently detected substances (Li et al. 2011; Arbuckle et al. 2013; Bao et al. 2014; Zeng et al. 2015; Lorber et al. 2015), with both PFOA and PFOS having an estimated 1000 days residence time in human blood (OECD 2013). Nevertheless, uncertainties remain among scientists as to what the possible health effects on humans exposed to PFCs could be, since, of the PFCs that have been found to accumulate in the human body, the levels of accumulation have been seen decreasing slowly over time (ATSDR 2015, 2016). Conversely, available data have indicated that the ability of PFCs to be bioaccumulate in the human body, also referred to as body burden, has increased concerns about the possibility of these compounds to cause detrimental health effects on human (ATSDR 2015, 2016). Hence, a number of human studies have reported that certain PFCs may affect foetus and child development, including retarding child growth, learning and behaviour (Ek et al. 2012; ATSDR 2015, 2016), while others have found inconsistent association between PFOA or PFOS serum levels and changes in reproductive hormone levels (Raymer et al. 2012; Specht et al. 2012; Joensen et al. 2013). On the other hand, conflicting results were found in studies investigating the association of sperm parameters (Toft et al. 2012; Raymer et al. 2012; Joensen et al. 2013) and impaired fertility (Fei et al. 2012; Vestergaard et al. 2012; Whitworth et al. 2012). Similarly, lines of evidence have further indicated that PFC, such as PFOA, exposure increases cholesterol (Frisbee et al. 2010; Eriksen et al. 2013) and affects

the immune system (ATSDR 2015, 2016). In addition, increases in prostate, kidney and testicular cancers have been reported in workers and communities living near PFC manufacturing facilities (ATSDR 2015). Nonetheless, there are limited data on whether PFC exposure can lead to cancer in humans, further suggesting that more research is still needed in this regard. Additionally, reproductive toxicity studies have also revealed possible associations between serum PFC levels and changes in reproductive hormone levels in men. Nevertheless, there have been inconsistencies in the reported results. For instance, Raymer et al. (2012) found significant positive correlations between PFOA levels and free testosterone and LH levels, but not with other reproductive hormones, while in a similar study by Joensen et al. (2013), no significant associations between reproductive hormone levels and serum PFOA, and other PFCs, such as PFHxS or PFHpS, were found. In contrast, no associations between serum PFOS levels and reproductive hormones were found by Raymer et al. (2012), while a significant negative correlation between PFOS and testosterone, free testosterone and free androgen levels was found by Joensen et al. (2013) in young men. Table S5 provides a brief toxicological summary of available epidemiological data present in the reviewed literature on reproductive effects in humans exposed to PFCs.

Furthermore, even though PFCs have been studied in a number of human epidemiological studies and their prevalence reported in human tissues, including blood samples (Genuis et al. 2013), there are still no reports of human deaths from accidental or intentional acute exposure to high concentrations of PFOA or PFOS (ATSDR 2015). However, most studies have indicated the potential associations between mortality and long-term exposure to these substances. For example, a study by Alexander et al. (2003) found no death increases from all causes led by PFOS, and Leonard et al. (2008) indicated the same from all PFOA causes.

Polyfluoroalkyl compound pathways into humans

The presence of chemical compounds in the environment does not automatically translate into human exposure. Typical exposure depends on a number of parameters, including, but not limited to, the degree of exposure. Thus, a growing body of evidence suggests that human exposure to PFCs and their potential precursors can be divided into three major categories, namely,

occupational exposure, general human exposure and exposure from mother to foetus or infants.

Occupational exposure

This form of exposure occurs during the performance of normal and legally delegated job requirements/responsibilities. Thus, workers in facilities that manufacture PFCs or in the formulation and production amenities that use products containing PFCs, direct exposure is through handling of such preparations, having contact with processing liquids, wastewater or treated products or when carrying out maintenance, sampling, testing or other procedures. For example, high levels of PFOS and PFOA were found in workers at PFC production sites (Freberg et al. 2010; OECD 2013).

General human exposure

A growing body of scientific evidence has also revealed that general human exposure to PFCs and its precursors occurs by way of (i) indoor and outdoor air and aerosols, (ii) contaminated drinking water, (iii) food and (iv) dust (D'Hollander et al. 2014, 2015; Pérez et al. 2014; Duong et al. 2015; Brambilla et al. 2015; Filipovic et al. 2015; Koponen et al. 2015; Liu et al. 2015; Schlummer et al. 2015). Accordingly, PFCs and their precursors can be found in various food items (Post et al. 2012; Yeung et al. 2013; OECD 2013). In addition, it has been argued that exposure via dust particles might be a minor exposure pathway for adults in comparison to dietary intake (Xu et al. 2013; OECD 2013), although it may be a significant pathway for infants and toddlers (Fromme et al. 2009; D'Hollander et al. 2010; OECD 2013). Overall, tap water and agricultural produce, irrigated with contaminated river water, have all been found to be a significant source of exposure for humans (Tabtong et al. 2015; Chen et al. 2016; Hurley et al. 2016). Recent research has indicated that paper and packaging for food, as well as different materials used for food contact, play a contributory role in the contamination of food from PFCs (Surma et al. 2015; Shoeib et al. 2016). Table 2 depicts evidence of PFCs containing wrappers from different food contact paper, food brands and beverages.

Foetal and/or infant exposure to PFCs

The exposure of a foetus and/or infants to PFCs has been of particular concern and is not well understood.

Table 2 Evidence of PFC content in fast-food wrapper (Schaidler et al. 2017)

	Brands tested (<i>n</i>)	Samples tested (<i>n</i>)	PFC content (%)
Food contact wrapper (by type)			
Sandwich/burger	20	138	38
Dessert/bread	9	69	56
Tex-Mex	3	42	57
Food contact wrapper (all)	27	248	46
Food contact paperboard	15	80	20
Non-contact paper	9	15	0
Paper cups	9	30	0
Other beverage containers	10	25	16
Miscellaneous	7	9	0

This group has a higher risk of PFC exposure (Fromme et al. 2009; OECD 2013). However, from mammalian studies, it is known that PFCs are able to transcend the placenta and enter the foetus (Gützkow et al. 2012). From a human perspective, it is suggested that this exposure occurs in two ways, namely, (i) through the placenta to the foetus (Cariou et al. 2015) and (ii) from lactating mothers to their infants through breast-feeding (Mogensen et al. 2015; Kang et al. 2016).

However, Fromme et al. (2009) have argued that the mechanism by which PFCs are transferred from the mother's blood to breast milk has remained unclear, although further evidence has suggested that PFCs are strongly bound to the protein fraction in the blood (Han et al. 2003; Li et al. 2013). In addition, it was previously reported that, PFC, that is, PFOA, levels in maternal blood decreased from 54 to 7% after 6 and 12 months, of breast-feeding, respectively, compared to their levels in blood at child birth (Thomsen et al. 2010), while PFOA levels in the serum of 6-month-old infants were 4.6 times higher than maternal blood levels at birth (Fromme et al. 2010), suggesting that further exposure pathways had contributed to the sudden increase. Similarly, breast-fed infants of around 6 months of age take up 4.1 ng kg⁻¹ bw day⁻¹ of PFOA, which is 15 times higher than the uptake in adults (Haug et al. 2011). The question is "Did age-related exposure play a role in this instance?" It is unclear at this point simply because the largest part of studies that have studied the correlation of age with PFC concentrations in blood has not observed significant effects (Calafat et al. 2007; Fromme et al. 2009), although PFCs such as PFOA and PFOS are

POPs which do not biodegrade. It might be expected that the BMF would rise with age, just as it was reported with other POPs in Duarte-Davidson and Jones (1994) and Knowler et al. (2014).

Toxicity and health risks associated with perfluoroalkyl compounds

The toxicity of PFCs differs from other POPs, and this has led to their toxicokinetic mechanism being unknown (Senevirathna 2010). Nevertheless, medium- and long-chained PFCs are believed to be more toxic than short-chained PFCs (Renner 2006; Senevirathna 2010). Accordingly, both PFOA and PFOS seem to be readily absorbed through oral intake (that is ingestion or gaseous) but are poorly eliminated from the human body (Lau et al. 2007; Møskeland 2010). Both PFOA and PFOS do not biodegrade substantially, due to their stability, and thus, tend to accumulate into the endocrine system, for example, the kidney, liver or other organs possibly, in the case of the liver, as a result of attaching to certain proteins, such as β -lipoproteins, albumin and fatty acid binding proteins in the liver, as it has been demonstrated to be the primary beleaguered organ by PFCs (Fang et al. 2015; Midgett et al. 2015; Li et al. 2016). To elaborate on this, PFCs have previously been regarded as peroxisome proliferators (PPs), suggesting that they can lead to a variety of toxicological effects on the liver, including carcinomas (Vaughn et al. 2013; Krafft and Riess 2015). PPs include certain hypolipidaemic drugs, phthalate ester plasticisers, industrial solvents, herbicides, food flavourings, leukotriene D4 antagonists and hormones (Reddy 2004). Furthermore, PFOS and PFOA have half-lives in humans ranging from 2 to 9 years, but it has been argued that this half-life coupled with continued exposure can increase the burden of the human body and ultimately lead to levels that would result in adverse outcomes over the long term (EPA 2014; ATSDR 2015). Chronic toxicity reports have associated PFOA exposure with tumours (Rosen et al. 2009; Wan et al. 2013), while severe and intermediary duration oral studies on rodents have indicated risks associated with potential stunted developmental, reproductive and other systemic growth defects (EPA 2014). It was also suggested that PFOA and PFOS are able to compete with thyroxin which is linked with the human thyroid hormone transport protein transthyretin (Weiss et al. 2009; Møskeland 2010). In general, this

appears to be the effect of longer-chained PFCs than shorter-chained PFCs (for example, PFBS). This finding has prompted a shift in industry practice to favour shorter-chain PFCs (Renner 2006), which is detrimental to the efforts to eradicate PFC usage worldwide (Jensen and Warming 2015). Table 3 depicts a brief summary of the results from various studies on PFCs' toxicities in wild lives.

Moreover, recent studies have demonstrated that PFCs may induce reactive oxygen species generation and induce deoxyribonucleic acid (DNA) damage in the cells of humans and livers of wildlife animal (Reistad et al. 2013; Mashayekhi et al. 2015). Additionally, in a retrospective cohort mortality study in which more than 6000 PFOA-exposed employees were involved, results reported elevated standardised mortality ratios for kidney cancer, as well as a significant increase in diabetes mortality for male workers, although the study indicated that further investigations were required to substantiate its findings (Lau et al. 2007; EPA 2014). Evidence from Melzer et al. (2010) and White et al. (2011) also reported that higher concentrations of PFOA and PFOS in human sera were associated with thyroid disease in elderly persons. However, the study suggested that further analysis was required to identify the mechanisms allowing this association (Melzer et al. 2010).

In addition, PFOS exposure was also associated with bladder cancer (Chang et al. 2014; Grandjean and Clapp 2015). In vitro and in vivo epidemiologic and immunotoxicologic studies reported that high levels of PFCs in adults and children correlated with observed changes in their health, including decreases in IgE levels, coupled with increases in antinuclear antibodies, asthma, influenza and gastroenteritis (Keil 2015). To mitigate the health effects associated with long-chain PFCs, it was suggested that alternative commercially available short-chain compounds should replace these compounds (Poulsen et al. 2005).

Commercially available alternatives to long-chain perfluoroalkyl compounds

For decades, long-chain PFCs, including PFOA and PFOS, were used in various industrial applications (Wang et al. 2014a, b; Taniyasu et al. 2015; Niu et al. 2016). However, concerns over the effect of these compounds on humans and the environment led to an interest in exploring suitable alternatives to these chemicals

Table 3 Brief summary data on PFOA and PFOS toxicities (Stahl et al. 2011)

Compound	Exposure time	Species type	Organ tested	Effect	Dosage	NOAEL	Reference
PFOA	7 days	Japanese guppies	n.i.	Activity of peroxisomal acyl-CoA-oxidase ↑	2 to 20 mg/kg feed	n.r.	Yang (2010)
	14 days	Minnows	n.i.	Changes in the expression of Apo lipoproteins and upstream genes	n.r.	n.r.	Fang et al. (2010)
PFOS	90 days	Rats (male)	Liver	Liver mass ↑ and hepatocellular necrosis	1.7	0.6	Cui et al. (2009)
	28 days	Rats	Liver and other	Body weight ↓, liver mass ↑ and altered gene expression and fatty acid metabolism in the liver, T ₃ and T ₄ ↓	2 to 20 mg/kg feed	n.r.	Curran et al. (2008)
PFOS	14 weeks	Rats (male)	Liver	Hypertrophy and vacuolization of the liver	n.r.	0.37	Seacat et al. (2003)
	26 weeks	Cynomolgus monkey	Liver and other	Centrilobular vacuolization, hypertrophy of the liver, T ₃ ↓, TSH ↑, HDL ↓, and bilirubin, cholesterol concentrations ↓	n.r.	0.03	Seacat et al. (2002)
	1 and 4 months	Fresh water larvae	n.i.	Deterioration of behavioural and activity parameters (larvae were less active, less able to avoid attackers, or less efficient in foraging)	>10 µg/L	10 µg/L	Van Gossum et al. (2009)

T₃ tri-iodo thyronine, T₄ thyroxine, upward arrow increased, downward arrow decreased, n.r. not reported, n.i. not indicated

(Jenssen et al. 2015). Thus, there are three types of available alternatives to long-chain PFCs, namely, (i) substances with shorter per- or polyfluorinated carbon chains, (ii) non-fluorine-containing substances and (iii) non-chemical techniques (OECD 2013).

Substances with shorter per- or polyfluorinated carbon chains

The discontinuity of the manufacturing of “C₈-chain” fluorinated compounds was agreed upon between the manufacturers of these chemicals and regulatory agencies (for example, the Stockholm Convention on POPs) decades ago. Hence, equivalent “short-chain” fluorinated substances were suggested as alternative replacements, with indications suggesting that they were less hazardous and can be manufactured as substitutes for applications in which long-chain PFCs were used (Holt 2011; OECD 2013; Jenssen et al. 2015). Thus, examples of suggested replacement compounds included (i) 6:2 fluorotelomer-based chemicals, (ii) perfluorobutane sulfonyl fluoride (PBSF)-based derivatives, (iii) mono- and polyfluorinated-ether-functionality compounds, (iv) fluorinated oxetanes; and (v) other fluorinated polymers (Buck et al. 2011; OECD 2013).

Furthermore, it has been indicated that the most important short-chain PFCs were perfluorobutane sulfonate (C₄, PFBS) and perfluorohexane sulfonic acid (C₆, PFHxS) (Jenssen et al. 2015). Table 4 depicts some of the commonly known commercially available short-chain alternatives.

Non-fluorine-containing substitutes

Non-fluorine-containing compounds with similar properties to those seen in PFCs are available commercially and some have been used in various industrial applications (OECD 2013), namely, (i) naphthalenes or biphenyls used as water-repelling agents for rust protection systems, marine paints and coating, among others; (ii) fatty alcohol polyglycol ether sulphate used as a leveling and wetting agent; (iii) sulfosuccinates used for surface coating, paints and varnish; (iv) hydrocarbon surfactants used in the photographic industry; (v) siloxanes and silicone polymers used for impregnation of textiles, leather and carpets; (vi) stearamidomethyl pyridine chloride which is also used for impregnation of

textiles, leather and carpets; and (vii) polypropylene glycol ether, amines and sulphates. However, it has been noted that these alternatives may have limited usability when compared to their long-chain predecessors (Holt 2011; OECD 2013). Conversely, some of these alternatives have been determined to be hazardous to humans (Dong et al. 2013; Gorrochategui et al. 2014), although conclusive results are still required. In addition, critics suggest the health and environmental profiles of these substitutes to be fully tested before their large-scale commercialisation.

Potential health impact associated with short-chain perfluoroalkyl compound alternatives

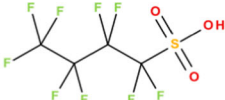



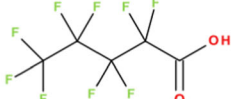
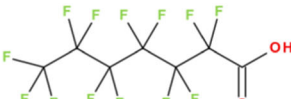
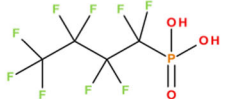
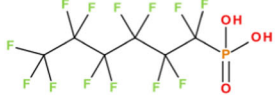


Firstly, a recent study has indicated that various known short-chain PFCA and PFSA have similar physico-chemical properties as those seen in long-chain PFCs, such as high water solubility and persistency, among others (Gomis et al. 2015). Two decades ago, a trend driven by concerns over long-chain PFCs and their undesirable impact on humans and environmental health resulted in the development of alternative compounds worldwide among PFC producers, in order to replace C₈-fluorocarbons (Wang et al. 2013).

However, information on their impact, including their bioaccumulative potential in the environment, has generally remained limited and is not readily available (Wang et al. 2013). The OECD (2013) has indicated that this lack of information has been due to confidentiality and trade secret concerns, while Wang et al. (2013) have argued that these alternatives to long-chain PFCs applied similar production techniques such as polymerisation which suggested that they may enter the environment, including surrounding production sites where they were produced and used, which, in the long term, will mimic similar distributary mechanisms observed for long-chain PFCs.

Accordingly, various studies have reported short-chain alternatives to PFCs in several matrices using similar research techniques to those applied for long-chain PFCs. For instance, elevated levels of PFBS and other precursors have been detected in water samples in Germany (Möller et al. 2010), Japan (Ahrens et al. 2010) and the Northwest Pacific Ocean (Cai et al. 2011).

Nevertheless, published research has argued that due to concerns over intellectual property rights, required data to assess the safety of these substitutes has not yet been established (OECD 2013; Wang et al. 2013). The

Table 4 Some of the commonly known commercial alternatives to long-chain PFCs (Jenssen et al. 2015)

Compound and Acronyms	Chemical structure
<i>Perfluorobutane sulfonic acid (PFBS)</i>	
<i>Perfluorohexane sulfonic acid (PFHxS)</i>	
<i>N-Methyl perfluorobutane sulfonamidoethanol (MeFBSE)</i>	
<i>N-Methyl perfluorohexane sulfonamidoethyl acrylate</i>	
<i>Perfluorobutanoic acid (PFBA)</i>	
<i>Perfluorohexanoic acid (PFHxA)</i>	
<i>Perfluorobutyl (PFBPA)</i>	
<i>Perfluorohexyl phosphonate (PFHxPA)</i>	
<i>4:2 Fluorotelomer alcohol (4:2 FTOH)</i>	
<i>6:2 Fluorotelomer phosphate/mono[2-(perfluorohexyl)ethyl] phosphate</i>	

lack of such information has made it possible for critics to question whether these alternatives have been fully scrutinised prior to their commercialisation (Wang et al. 2013). There has been no focus on the environmental health impact of PFC substitutes in countries with lower or non-existent regulatory requirements; therefore, regulatory monitoring and reporting mechanisms are non-existent even for long-chain PFCs, for example, in South Africa. This reality has further inhibited researchers, regulators and other civil society stakeholders from assessing and developing strategies that can minimise the risks associated with these substitutes, without monitoring activities and studies into the environmental fate and potential adverse effects of PFC substitutes. It is therefore difficult to mitigate their impact in the long term (Goldstein et al. 2013; Wang et al. 2013).

There are suggestions which indicate that short-chain PFC alternatives are less bioaccumulative (Wang et al. 2013) and toxic (Borg and Hakansson 2012), although recent scientific evidence has suggested that short-chain PFCs have shown a higher uptake into the leaves, stems and fruits of plants (Krippner et al. 2014, 2015). This ultimately suggests that these contaminated floras will constitute a major pathway into humans. Among the PFC alternatives, that is, PFBS, PFBA, PFHxS and PFHxA (Krippner et al. 2015), PFBS has been shown to be persistent in the environment, a characteristic observed in their C₈-homologues (Wang et al. 2013). Although PFBA, PFHxS and PFHxA, including PFBS, have shorter half-lives in both humans and biota than their longer-chain homologues (Iwai 2011; Borg and Hakansson 2012), current studies have reported that some PFHxA can even have longer serum half-lives than long-chain PFCs, such as PFOS, suggesting the unsuitability of using these compounds as alternatives (Wang et al. 2013).

Additionally, the Asahi Glass Company (2006) described PFHxA as being acutely toxic, three to five times more than PFOA, with PFBS being reported to cause disruptive effects on cell membranes (Oldham et al. 2012; Jensen and Warming 2015) and having the potential to act as an aromatase inhibitor in placental cells (Gorrochategui et al. 2014). PFBS and PFHxS have been suggested to have an effect on how lipids are metabolised (Bijland et al. 2011; Jensen and Warming 2015). Thus, PFHxS leads to liver weight increases. Relevant data on the content of short-chain PFCs in human organ tissues and PFOA/PFOS are shown in Table 5. Hence, in order to reduce the potential

impact of both long-chain and their suggested substitutes, some novel technologies have been developed for either the decomposition and/or treatment of these compounds, particularly at the point of use. Currently, these technologies are still at the laboratory level and have yet to be implemented on a large scale.

Some of the novel technologies used for the treatment and/or removal of polyfluoroalkyl compounds in water

Concerns over the prevalence of PFCs in the ecosystem have increased during recent decades. However, the treatment and removal of these compounds from contaminated water have remained a challenge. The unique physicochemical properties, including strong fluorine-carbon bonds in PFCs, have contributed to these compounds being resistant to most conventional treatment technologies (Arvaniti and Stasinakis 2015).

Currently, advanced treatment technologies have emerged with regard to reduction processes and advanced oxidation (Arvaniti and Stasinakis 2015), including electrochemical treatment (Schaefer et al. 2015), processes which have been proven to be suitable for the treatment of PFCs in environmental matrices. Furthermore, treatment at the point of use can be harnessed to reduce PFCs. Some well-established PFC treatment/removal processes include the use of adsorption and advanced membrane filtration systems. Overall, all these processes are designed for the treatment of potable water and wastewater.

Granular activated carbon adsorption

For well-established processes, adsorption has been the most common remediation technology used for PFCs, which is based on PFC adsorption into GAC (Shih and Wang 2013; Arias-Espana et al. 2015). Thus, four steps, namely, (i) diffusion from the liquid phase, (ii) mass transfer on to the solid phase, (iii) internal diffusion (pore and surface diffusion) inside an adsorbent and (iv) electrostatic and/or hydrophobic interaction with the exchange site, were identified by Yong (2007) as being critical in the adsorption mechanism using activated carbon. Thus, Vecitis et al. (2009) reported that GAC is utilised to remove PFCs, in this case PFOA and PFOS, and has been proven effective in removing both substances at more than 90% mass of PFC removal/

Table 5 Concentration of short-chain PFCs in five human organ tissues (Pérez et al. 2013; Jensen and Warming 2015)

PFC substance	Mean concentrations ng/g w.w.				
	Liver	Bone	Brain	Lung	Kidney
PFBS	0.9	3.2	<LOD	17.8	8
PFBA	12.9	<LOD	13.5	304	464
PFPeA	1.4	0.8	<LOD	44.5	<LOD
PFHxA	11.5	35.6	18.0	50.1	5.6
PFHxS	4.6	1.8	3.2	8.1	20.8
FHEA (metabolite of 6:2 FTOH)	92.6	42.5	18.6	2.4	23.7
PFOA	13.6	60.2	<LOD	29.2	2.0
PFOS	102	<LOD	4.9	29.1	75.6

LOD limit of detection, w.w. wet weight

mass of GAC used (mg/g GAC) subsequent to the thermal treatment of GAC, with results indicating minimal residual PFC post-thermal treatment (Watanabe et al. 2015). However, controversial views have been raised in the literature on the ability of GAC to remove PFOS and PFOA. For instance, although GAC has been demonstrated to remove PFOS at microgram per litre levels, this is not the case for PFOA (Senevirathna et al. 2010; Appleman et al. 2013). Several other studies have indicated that factors such as carbon-fouling and pre-washing, as well as the presence of organic matter and high salinity, can decrease PFC removal which affects adsorption and the modification of surface properties of the GAC (Yu and Hu 2011; Appleman et al. 2013). Additionally, Hansen et al. (2010) have indicated that commercial GAC has been mainly used to investigate PFOS and PFOA removal, with the removal of other PFCs, including the proposed short-chain substitutes remaining unknown.

Recently, GAC/PFC removal has also been achieved using natural sources such as Bambusoideae (bamboo) and *Agave sisalana* (Deng et al. 2015a; Mudumbi et al. 2015). Furthermore, various other adsorbents have been utilised in the treatment and removal of PFCs and have included powdered activated carbon (PAC), carbon nanotubes, mesoporous carbon nitride commercial resins, polymers, maize straw-derived ash, alumina, chitosan, goethite, silica, montmorillonite, organo-clay, hexadecyltrimethylammonium bromide (HDTMAB)-immobilised hollow mesoporous silica spheres, cetyltrimethyl ammonium bromide-modified sorbent, permanently confined micelle array (PCMA) sorbents and electrospun fibre membranes (Senevirathna et al.

2010; Hansen et al. 2010; Zhou et al. 2010b; Tang et al. 2010; Deng et al. 2010, 2012, 2015b; Yu and Hu 2011; Chen et al. 2011; Wang and Shih 2011; Zhang et al. 2011; Das et al. 2013; Zhou et al. 2013; Dai et al. 2013a, b; Xu et al. 2013; Yan et al. 2013; Bei et al. 2014; Chularueangaksorn et al. 2014a; Yao et al. 2014; Li and Zhang 2014; Wang et al. 2014c). However, when comparing PAC and GAC, evidence has reported higher and faster removal of PFOS and PFOA using PAC rather than GAC (Arvaniti and Stasinakis 2015), whereby the adsorption equilibrium was reached in 6 h during PAC treatment, which escalated to 168 h during GAC treatment (Senevirathna et al. 2010; Arvaniti and Stasinakis 2015). A similar trend was also reported by Arias-Espana et al. (2015). This suggests that exchange sites in PAC are more suited to PFC removal than those in GAC. Therefore, ion/site exchange effectiveness can effectively determine the success of a treatment strategy and thus the development of resin-based treatment methods.

Anion resin ion exchange adsorption

Numerous studies have indicated the suitability of ion exchange for the removal of pollutants (Alesi and Kitchin 2012; Shkolnikov et al. 2012). According to Helfferich (1962), ion exchange resins are the most important class of ion exchangers, thus can be used to adsorb POPs.

It has been reported how ion exchange resins can be utilised to exchange unwanted ions with hydrogen or hydroxyl group to remove contaminants, including PFCs (Deng et al. 2010; Senevirathna et al. 2010;

Alesi and Kitchin 2012; Shkolnikov et al. 2012; Chularueangksorn et al. 2014b). It was reported that an anion exchanger was better than GAC in the removal of PFOA (Chularueangksorn et al. 2014b), while Appleman et al. (2014) demonstrated the effectiveness of an anion exchange in removing PFOS (>92%), PFOA (74%) and PFNA (>67%).

Nevertheless, regardless of the success of ion exchange resins, Chularueangksorn et al. (2014b) have indicated that resins are expensive. Thus, the report suggests that they should be periodically regenerated for re-use in the removal of PFCs. This suggestion, however, did not consider the effect of cross and cumulative contamination as some resin beads may contain residual PFCs even after regeneration. Additionally, it has been reported that the rate of removal using an anion exchange treatment is largely dependent on the concentration level of the contaminant, the concentration of competing ions and the treatment system design (that is, flow rate and the size of the resin bed) and the nature of the exchange ions within the resin (ITRC 2008; Cummings et al. 2015). Additionally, Appleman et al. (2014) and Rahman et al. (2014) have recommended that further research is needed to effectively comprehend and identify the most suitable resins for removal of various pollutants in general and PFCs in particular. These studies also noted that it is necessary to frequently change the resins to completely eradicate residual PFCs in the beads. The ITRC (2008) has further suggested that both the management of the resin and that of the brine should also be taken into consideration when anion resin is used.

Removal of PFCs by combination of adsorption and coagulation

Coagulation has been reported as another technique that can be utilised for the removal of PFCs. However, its efficacy has been questioned in most cases. For instance, no removal occurred even after coagulation processes were coupled with sedimentation and sand filtration in a study by Takagi et al. (2011). This was consistent with the results which were observed by Thompson et al. (2011), Eschauzier et al. (2012) and Xiao et al. (2013). Similarly, Appleman et al. (2014) further indicated that coagulation followed by sedimentation did not remove PFCs, but when sedimentation was replaced by dissolved air flotation (DAF), a 49% removal of PFOS was achieved, although shorter-chain PFCs, such as PFCAs and PFSAs, were not well removed (Appleman et al. 2014). This suggests that coagulation on its own is likely not to yield positive results. Thus, a study by

Deng et al. (2011) found that coagulation can remove most PFOA from water, but high residual PFOA concentrations remained in the water. In this regard, the study combined adsorption and coagulation and the removal was enhanced. Similarly, recent evidence has reported that the combination of adsorption by PAC and coagulation increased the removal ratios up to >90% for PFCs, such as PFOX with an initial concentration of 1 mg/L (Bao et al. 2014). Hence, this further implies how adsorption enhances coagulation. Nevertheless, it has further been indicated that in a PFC-adsorption technique where fulvic acid (FA) is used, its concentration (i.e. FA) increase decreases the removal ratio of PFOS and PFOA, simply due to the steric hindrance effect of this acid's molecules and the competitive adsorption of these PFCs (Bao et al. 2014), suggesting that the selection of coagulants, as well as that of adsorbents to be used during the coagulation/adsorption technique, etc., is also paramount. Hence, Du et al. (2014) reviewed PFC removal using various adsorbents and reported that adsorption not only removed PFCs effectively but also affected PFC distribution in different environments. However, Du et al. (2014) have argued that on the basis of C-F chain substances having hydrophobic and oleophobic properties, this implies that PFCs likely display different adsorption behaviours as compared to their counterparts, e.g. the hydrocarbon substances. Thus, the authors have suggested that aspect, coupled with the competitive adsorption of PFCs with other traditional POPs present in various environments, to be investigated (Du et al. 2014; Bao et al. 2014).

Nevertheless, the stubbornness of shorter-chain PFCs in resisting removal, as indicated by Appleman et al. (2014), remains a cause for concern, particularly, since there is not enough data available reporting on these new emerging kinds of POPs, even though their use as substitutes to long-chain PFCs is increasing (Rahman et al. 2014). This suggests that improved removal techniques for shorter-chains PFCs are required. On the other hand, Yang et al. (2016a, b) have suggested that to improve scaling-up PFC removal techniques, more understanding of the mechanisms that have been proven effective is required, as well as testing these mechanisms on various PFCs.

Advanced filtration: membrane-based treatment processes

Filtration has been broadly defined as a technique that separates suspended particles from a liquid phase by

causing the latter to pass through a porous filter, with the purpose of either removing the impurities and/or collecting them from the solution where they are concentrated (Crittenden et al. 2012). In the case of PFCs, sand filtration cannot be used for the removal of PFCs (Takagi et al. 2011; Eschauzier et al. 2012; Arvaniti and Stasinakis 2015). However, most potable water treatment works in developing countries, such as South Africa, still use sand filters. Conversely, it was reported that the usage of advanced filtration techniques such as nanofiltration (NF) and reverse osmosis (RO) achieved a significant reduction of PFCs (Schröder et al. 2010; Appleman et al. 2013; Stasinakis et al. 2013).

Nanofiltration

Introduced during the late 1980s (Mohammad et al. 2015), NF is another form of membrane technology process used with the purpose of softening and removing synthetic POPs (Rahimpour et al. 2010). Thus, Izadpanah and Javidnia (2012) have indicated that this method of filtration provides high water flux at low operating pressure. It has been shown that NF can be effective in the removal of PFCs. Similarly, Tang et al. (2007) and Schröder et al. (2010) reported 90 and 99% removal of PFCs using NF. However, lower removal rates (that is, 44 to 86%) were reported by Rattanaoudom (2011), suggesting that the technique is inefficient. As such, Arias-Espana et al. (2015) indicated that pH is an important factor that affects nano-membrane retention rates for POPs. Similarly, at a $\text{pH} \leq 3$, Steinle-Darling and Reinhard (2008) and Wang et al. (2015a, b) observed a decline in the rejection of PFC (35%), and Wang et al. (2015a, b) also observed that PFOS rejections improved from 91.17 to 97.49% with an increase in pH from 3.2 to 9.5 at 4×10^5 Pa. However, a similar study reported that PFOS removal using NF was higher than for PFOA (Rattanaoudom 2011), a result which was also observed by Yu et al. (2014) with a removal efficiency of 77.4% for PFOS and 67.7% for PFOA. Additionally, Appleman et al. (2013) observed a 93% removal for all target PFCs through the usage of NF.

Moreover, recent research has focused on ways of improving NF effectiveness by modifying membrane materials used, with the purpose of increasing the strength, heat resistance, functionality and other factors (Luo et al. 2016). As such, several inorganic fillers, for example, zeolites (Gevers et al. 2005), ceramic oxides

(Pages et al. 2013; Schmidt et al. 2014; Zhang et al. 2014), inorganic compounds (Fang and Duranceau 2013; Namvar-Mahboub and Pakizeh 2013; Gholami et al. 2014; Chen et al. 2014) and layered silicates have been used. The reason being that their dispersion is possible in polymeric matrices at the nanoscale (Luo et al. 2016), which can further enhance membrane electro-chemical properties which are essential in filtration systems, particularly for the removal of compounds with unique properties, such as PFCs, compounds containing a hydrophobic backbone and hydrophilic functional groups.

Reverse osmosis

RO, as a POP treatment process, uses high pressure to force water through a semi-permeable membrane (Lee et al. 2010). Hence, Letterman (1999) indicated the removal of salts from brackish water and seawater, as the primary usage of RO, although the same technique can also be used for high rejection of synthetic organic compounds (SOCs), such as PFCs. Thus, Vecitis et al. (2009) reported that RO has shown its effectiveness in PFC removal. Another study showed $\geq 99\%$ removal of PFOS and PFOA (Flores et al. 2013). Similarly, it was revealed in a study by Tang et al. (2007) that RO had a higher efficacy in PFC removal than NF. This was attributed to the smaller pores and thicker rejection layers of the RO membranes used. In a hybrid membrane experiment where the reduction of turbidity from fire-fighting foam wastewaters was used, a 71 to 77% removal of fluorinated surfactants was reported. However, from a pilot fire-fighting foam wastewater treatment plant where RO was used, rejection rates $>99\%$ were achieved (Baudequin et al. 2011; Arias-Espana et al. 2015).

Nevertheless, regardless of the high efficiency of the RO, criticism about its use is based on the relatively high operational costs associated with the technology due to energy-intensified requirements of the system (Joo and Tansel 2015). Additionally, it also has been indicated that the RO is susceptible to biofouling, for which an improvement is required to enhance its usability in communities with minimal investment capital (Henthorne and Boysen 2015).

Furthermore, recent evidence indicates the versatility of RO systems and their effectiveness in new applications with proponents suggesting that RO can outperform other desalination technologies (McGovern 2014).

As such, forward osmosis (FO) has been investigated in the past decade, not to replace RO but to be utilised to process feed waters that cannot be treated by RO (Shaffer et al. 2015). This further suggests that, to date, there is no generally accepted technique which is readily available for the removal of PFCs and other perfluoroalkyl pollutants. Ultimately, the degradation and/or decomposition of PFCs might be the only viable option, with advanced oxidation processes having been reported to be suitable.

Advanced oxidation processes

According to Arias-Espana et al. (2015), the chemical structure of PFCs, mostly PFOA and PFOS, allows them to resist oxidation owing to the complete substitution of hydrogen (C-H bond) for fluorine (C-F bond). Fluorine atoms resist oxidation because it is the most electronegative element. This has been explained by Wardman (1989) who argues that fluorine with a reduction potential of 3.6 V is thermodynamically unsuitable to be substituted with any other oxidant (Arias-Espana et al. 2015).

Furthermore, advanced oxidation processes (AOPs), coupled with hydroxyl radicals in combination with ozone (or O-atom), were determined to be suitable for the reduction of recalcitrant POPs (Arias-Espana et al. 2015). However, for POPs such as PFOA and PFOS, the AOP/OH/O₃ was determined to be ineffective, as PFOA and PFOS do not contain hydrogen atoms which can be reduced at pH values commonly prevalent in the ecosystem (Arias-Espana et al. 2015). Hence, Schröder and Meesters (2005) argued that compounds such as PFOA and PFOS become inert to advanced oxidation mechanisms due to the substituted hydrogen by fluorine atoms in these POPs. Moreover, in situ advanced oxidation has been explored as a possible mechanism to treat PFCs in the environment (Liu et al. 2012a b). As such, oxidation processes have on several occasions been tested against recalcitrant contaminants (Arvaniti and Stasinakis 2015), during which the in situ formation of highly oxidising species, mainly free radicals, was involved.

Therefore, it was suggested that a variety of reagents have to be supplemented in AOPs in an attempt to enhance these oxidation processes. These supplementary compounds include activated persulfate, Fenton's agent, subcritical water, zero-valent metal and/or a combination of these agents (Arias-Espana et al. 2015). Supplementation with hydrogen peroxide (H₂O₂) has

been commonly used, due to its capability to generate hydroxyl radicals (HO^{*}), as well as persulfate (S₂O₈²⁻), Fenton's reagent (Fe²⁺ + H₂O₂) (Rayne and Forest 2009) and peroxymonosulfate (HSO₅⁻) (Antoniou and Andersen 2015; Arvaniti and Stasinakis 2015).

Hydrogen abstraction allows hydroxyl radicals to attack the organic substances by forming carbon centre radicals during the oxidation processes (Antoniou and Andersen 2015). Thus, because of the nonexistence of hydrogen atoms in PFCs that can be abstracted, this limits hydroxyl radicals' ability to react with these POPs, reducing the direct electron transfer (Vecitis et al. 2009; Arvaniti and Stasinakis 2015).

Additionally, a significant number of photolytic methods have been reported to effectively degrade PFCs into fluoride ions, carbon dioxide and shorter-chain PFCAs in aquatic samples (Arvaniti and Stasinakis 2015). Photolytic methods such as H₂O₂ photolysis and photocatalysis (Hori et al. 2004), direct photolysis (Chen and Zhang 2006; Yamamoto et al. 2007), persulfate photolysis (Hori et al. 2005; Chen and Zhang 2006), alkaline isopropanol photolysis (Yamamoto et al. 2007) and photo-Fenton (Hori et al. 2007; Wang et al. 2008; Tang et al. 2012) are examples which can be used for PFC reduction. New methods have emerged such as thermal- or microwave-activated persulfate oxidation (Liu et al. 2012a), heat-persulfate oxidation (Hori et al. 2008; Rayne and Forest 2009; Lee et al. 2012) and ultrasonic treatment (Cheng et al. 2008; Lin et al. 2015a, b). These methods have been applied and proven to be effective in degrading PFCs. Thus, Hori et al. (2005) and Wang et al. (2010) revealed that the usage of persulfate produced highly oxidative sulphate radical anions (SO₄^{*-}) which significantly degraded PFOA to F⁻ and CO₂ as major by-products. However, it was reported that shorter-chain PFCAs were formed, that is, compounds which were suggested as replacements for long-chain PFCs, suggesting the inadequacy of the method. This inadequacy suggested that a secondary treatment stage is required. Similarly, PFOA degradation was achieved using a photocatalytic AOP persulfate at 50 mM [S₂O₈]²⁻ and a 4-h irradiation with PFOA at a concentration being 1.35 mM (Arias-Espana et al. 2015).

Moreover, others have demonstrated that a sulphite/UV process was efficient in reductive degradation of PFOA (Song et al. 2013). Accordingly, 100% removal of PFOA and an 88.5% defluorination were completed after 1 h and a reaction time of 24 h, respectively, under

a nitrogen atmosphere. Similarly, the use of a UV-Fenton process achieved a 95% PFOA removal (Tang et al. 2012). Due to the success of these processes, other reductive processes such as zero-valent iron processes have been developed.

Reduction processes using zero-valent iron

Although the removal and/or treatment of PFCs by means of reduction processes using zero-valent iron (ZVI) has remained limited (Arvaniti and Stasinakis 2015), a study by Hori et al. (2006) has reported that a the partial degradation of PFOS by microsized ZVI coupled with high temperature (>250 °C) including pressure of up to 20 MPa can be achieved. Similarly, Lee et al. (2010) demonstrated that PFOA was susceptible to degradation up to 68 and 73% after 2 and 8 h, respectively, using persulfate activated by ZVI. In addition, a recent study by Arvaniti et al. (2015) investigated the removal and/or treatment of various PFCs in water using nanoscale ZVI (nZVI), using the nZVI uncoated and coated with Mg-aminoclay (MgAC). This method reportedly has PFC removal ability ranging from 30 to 96% (that is from 10 mg/L) under acidic conditions (pH = 3), low temperature (20 °C) and high doses of synthesised nanomaterial (1000 mg nZVI/L). According to Arvaniti and Stasinakis (2015), both sorption and degradation mechanisms are responsible for PFCs' removal when coated nZVI was used, a process used to achieve higher removal rates. In order to improve the effectiveness of processes using specialised materials such as ZVI, electrochemical cells can also be used.

Electrochemical treatment of polyfluoroalkyl compounds

Recently, the use of an electrochemical cell and a Ti/RuO₂ anode in laboratory experiments was assessed, demonstrating an increase in both PFOA and PFOS decomposition with increased current density (Schaefer et al. 2015). Thus, at a current density of 10 mA/cm², the electrochemical treatment rate of both PFOA and PFOS was 46×10^{-5} and 70×10^{-5} [(min⁻¹) (mA/cm²)⁻¹ (L)], respectively (Schaefer et al. 2015), with a defluorination ratio of 58 and 98% recovery for both PFOA and PFOS, respectively. Similarly, a study by Lin et al. (2012) investigated the electrochemical degradation of PFOA in aqueous solution over anodes, such as Ti/SnO₂-Sb, Ti/SnO₂-Sb/PbO₂ and

Ti/SnO₂-Sb/MnO₂. The results revealed a 98.8% degradation ratio of the substance (i.e. PFOA), with a 73.9% defluorination ratio, which is inconsistent with that of Schaefer et al. (2015). Nevertheless, both studies (i.e. Lin et al. 2012; Schaefer et al. 2015) have reported that short-chain PFCs remained recalcitrant to electrochemical degradation mechanism, suggesting a poor performance of the electrochemical treatment of PFCs as previously reported by Zhuo et al. (2011) and the need for an enhanced technology in this regard. In addition, previous studies which used this treatment method have indicated that the electrochemical treatment of PFCs can be efficient and yield significant results, in divided electrochemical cells rather than in undivided cells (Agladze et al. 2007; Schaefer et al. 2015). However, minimal research data are available in this regard, that is the evaluation of divided cells (Schaefer et al. 2015). The application of an inert environment, high temperature and pressure can further enhance electrochemical treatment.

On the other hand, electrocoagulation using stainless steel rod as cathode has recently emerged as an efficient PFC removal technique, by achieving a removal ratio of 99.7%/98.1% and 98.9%/97.3%, using stainless steel and aluminium rods as cathodes in the presence of different anions (e.g. Cl⁻/NO₃⁻), respectively (Wang et al. 2016). Previously, Lin et al. (2015a, b) demonstrated that hydrophobic interaction was a prime role player in PFC sorption and removal, a condition under which zinc anode proved to be more efficient than the other three anode materials, with 96.7% removal capacity. Hence, both these studies, i.e. Lin et al. (2015a, b) and Wang et al. (2016), are evidence that the electrocoagulation technique under various driving forces is an effective and alternative method to remove PFOA from aqueous solution. Nevertheless, it remains unclear what would be the removal effectiveness of this technique on short-chain PFCs. Similarly, different influencing factors, including pH, etc., can also be contributing factors in the removal of PFCs in various environments. Hence, Table S6 provides an overview comparison summary of results for PFC removal using different techniques.

Although technologically advanced, these methods require specialised knowledge, which limits practical application compared to cheaper options that rely on removal at the point-of-use (PoU).

Removal of PFCs at the point-of-use

This technique uses PoU treatment devices which are applied and/or installed at an individual or single tap, faucet or outlet for the purpose of reducing contaminants at that point-

of-use (Lee et al. 2005; MDH 2008). As such, a study by MDH reported that when applied, installed, operated and maintained according to the manufacturer's specifications, PoU treatment devices effectively remove PFCs (MDH 2008). In the report, it is suggested that devices were evaluated for their PFC removal capabilities, using an assessment classified into two categories, that is (i) those using GAC and (ii) those using a combination of multiple methods for the removal. From the results, it was revealed that some devices ($n = 11$) were found to remove PFCs in field tests to below the employed detection limits (50 ng/L) (MDH 2008). Additionally, in the late 1990s, a point-of-use plasma abatement (PPA) method was reported as one way to effectively eliminate PFCs at PoU (Fiala et al. 1999).

Conclusion

Perfluoroalkyl compounds (PFCs) are a group of chemical substances that fall under recalcitrant POPs. They consist of a fully fluorinated hydrophobic alkyl chain attached to a hydrophilic-end group. The unique physicochemical properties of these substances led to their extensive industrial and household applications, particularly in surfactants, fire-fighting foams and food-packing paper, as well as in textile, carpet and leather treatment. There are many types of PFCs, but the most widely used have included PFOA and PFOS. Recently, there have been studies reporting on PFBS as a potential replacement, as it has PFC characteristics and similar health risks as those associated with PFOA and PFOS. Thus, notwithstanding the role they have played in industrial and household applications, PFCs have been regarded as bioaccumulative, persistent and potentially precarious to humans and wildlife. For this reason, the development of alternatives to these compounds is underway. Ultimately, this has led various manufacturers to utilise short-chain PFCs in substitution of long-chain PFCs. However, like their homologues, short-chain PFCs have also been associated with various health risks. This finding suggests that further investigations are needed in this regard, since most studies have mostly focused on health-related risks of long-chain PFCs. To mitigate associated health risks to humans and animals, numerous treatment methods have been suggested, although treatment at point-of-use is currently the only viable option available to the general population. In our opinion, it is worth indicating that short-chain PFCs are recalcitrant, even to highly efficient removal techniques;

this is a challenge that requires the attention of researchers.

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