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Per- and polyfluoroalkyl substances (PFASs) in drinking water: current state of the science

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Abstract

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are ubiquitous emerging contaminants that have been used in many applications since the 1950s. They have been detected in many drinking water sources, sometimes at $\mu\text{g/L}$ level in water impacted by point sources. Following growing concerns on adverse ecological and human health effects, in the early 2000s several countries established regulations on the PFASs of most concern. Fluorinated alternatives were consequently developed by manufacturers, resulting in the release of novel PFASs that have recently been detected in water resources. Like legacy PFASs, most of them are recalcitrant to conventional drinking water treatments. Governments face the challenge of defining guideline values for water often containing a mixture of several PFASs with little conclusive toxicological and epidemiological evidence. Around 3,000 PFASs have been available on the global market, so the other key challenge is to identify the main ones in contaminated water resources, and to detect novel PFASs quicker than was the case for legacy PFASs.

Keywords

Per- and polyfluoroalkyl substances (PFASs); drinking water treatment; human exposure; point sources; guideline values

Introduction

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are a large group of persistent anthropogenic molecules containing the perfluoroalkyl moiety $\text{C}_n\text{F}_{2n+1}$ [1**,2*]. Perfluoroalkyl substances contain just a functional group (e.g. COOH , SO_3H) directly linked to the $\text{C}_n\text{F}_{2n+1}$ moiety. Polyfluoroalkyl substances contain the $\text{C}_n\text{F}_{2n+1}$ moiety, a functional group and at least one C-H bond in their chemical formula (e.g. $\text{C}_n\text{F}_{2n+1}\text{C}_2\text{H}_4\text{SO}_3\text{H}$, $\text{C}_n\text{F}_{2n+1}\text{SO}_2\text{NHC}_2\text{H}_4\text{OH}$). The difference in their behavior in the environment is huge, because perfluoroalkyl substances are extremely persistent, whereas polyfluoroalkyl substances have been shown to degrade into perfluoroalkyl substances under both biotic and abiotic conditions [1**].

Since the 1950s, PFASs have been used in a wide range of industrial and domestic applications, such as processing aids for fluoropolymer manufacture, surfactants in specific firefighting foams, and constituents of side-chain-fluorinated polymers for water- and grease-proof textiles and food containers [1**,3,4]. As a consequence of their widespread use and extreme resistance to

environmental degradation mechanisms, PFASs have been universally detected in many environmental compartments, including remote locations far from human settlements [1**,5*]. In the early 2000s, growing concerns about the ecological effects and human health impact of long-chained PFASs (with 7 or more perfluorinated carbons) have led to a drastic decline in their production and their progressive replacement by fluorinated alternatives with shorter chains, considered less bio-accumulative but unfortunately more mobile [1**,6]. Consequently, the phasing out and restricted use of long-chained PFASs will not lower the PFAS burden in the environment, but just result in different composition profiles being found in the environment.

Sources and occurrence in raw water

Of all PFASs, perfluoroalkyl carboxylic acids (PFCAs, $C_nF_{2n+1}COOH$) and perfluoroalkane sulfonic acids (PFSAs, $C_nF_{2n+1}SO_3H$) have been the most studied for various reasons, including the fact that i) they are highly persistent, ii) they have been directly released into the environment, and iii) they are the ultimate metabolites of many PFASs [1**]. In drinking water sources from non-industrial areas, PFCAs and PFSAs can be detected at the lower ng/L range due to transport over long distances and diffuse sources related to domestic applications [7-9]. However, the highest PFAS concentrations have been recorded near firefighter training areas [8-14], industrial sites making or using PFASs [11,15-21], landfill sites [22], and wastewater treatment plants [7,11] (Fig. 1).

Recent studies have emphasized that the analysis of PFCA and PFSA alone is insufficient to assess an environmental PFAS contamination from point sources. Besides legacy PFCAs and PFSAs, different emerging PFASs have thus recently been identified in the aquatic environment [5*], including drinking water sources (Table 1). However, probably at least 3,000 PFASs have been available on the global market [2*]. As it is impossible to individually identify and quantify all of them, novel analytic approaches such as the total precursor assay [23] or the total organic fluorine measurement [24] have been developed. They all aim to highlight the presence of unidentified PFASs in samples, but the interpretation of findings remains problematic in risk assessment terms [9].

Table 1: Emerging PFASs recently detected in drinking water sources and tap water

Name	Acronym	Formula	Concentration range (ng/L)		Reference
			Drinking water sources	Tap water	
Trifluoroacetic acid	TFA	CF_3COOH	<30-17,000	<30-11,000	J Janda, et al. [25]
Perfluoro-2-propoxypropanoic acid	PFPrOPrA (GenX)	$C_6HF_{11}O_3$	55-4,560	n.a.	M Sun, et al. [17*]
			n.a.	<0.2-11	WA Gebbink, et al. [18*]
Polyfluoroalkyl carboxylic acids		$C_nH_nF_nO_2$	n.a.	< 1 ^a	WA Gebbink, et al. [18*]
Polyfluoroalkyl sulfonic acids		$C_4H_2F_8SO_3$	n.a.	~ 1 ^a	
Mono-ether perfluoroalkyl carboxylic acids	moPFECAs	$C_nHF_{2n+1}O_3$	Detected but not quantified ^b		M Sun, et al. [17*]
Multi-ether	muPFECAs	$C_nHF_{2n-1}O_n$	Detected but not		

perfluoroalkyl carboxylic acids			quantified ^b		
6:2 Fluorotelomer sulfonic acid	6:2 FTSA	C ₈ H ₄ F ₁₃ SO ₃	0.32-11.4	n.a.	C Wei, et al. [7]
			<4-66	<4-16	X Dauchy, et al. [10], V Boiteux, et al. [16]
6:2 Fluorotelomer sulfonamide alkylbetaine	6:2 FTAB	C ₁₅ H ₂₀ F ₁₃ N ₂ SO ₄	<25-84	<25-32	
Chlorinated perfluoroalkyl ether sulfonate	6:2 Cl-PFAES (F-53B)	C ₈ ClF ₁₆ SO ₄	0.17-2.63	n.a.	C Wei, et al. [7]

^a Estimated concentrations based on structurally similar standards

^b 1 moPFECA and 2 muPFECAs exhibited peak areas 2-113 times greater than PFPrOPrA (GenX)

Removal efficiency of water treatments and PFAS concentrations in tap water

Several different PFASs are often found together in raw water samples due to the PFAS mixtures used in certain applications, fluorinated impurities contained in products and by-products of environmental transformation [15]. Water treatment technologies therefore have to remove a suite of PFASs with different and specific molecular properties (hydrophobic perfluorinated carbon tail of variable length linked to an anionic, neutral, cationic or zwitterionic functional group).

Traditional treatment technologies (i.e. coagulation, flocculation, sedimentation, sand filtration, biofiltration or UV irradiation) have been shown to be inefficient at removing PFASs [26] (Fig. 1). Granular activated carbon (GAC) filtration can be effective for long-chained PFCAs and PFSAAs when the GAC is new. Its removal efficiency depends on the contact time and water quality (competitive effects of ions and dissolved organic carbon) [26-28], and thus requires frequent replacement to avoid breakthrough [29,30]. The removal efficiency of powder activated carbon (PAC) is better than that of GAC because of its smaller particle size, resulting in a larger specific surface area and better accessibility to functional groups [26,30]. Anion exchange resins are more efficient than GAC in terms of total PFAS retention [27-30]. Furthermore, unlike GAC, some of them have the advantage of being easy to regenerate in situ [29]. High-pressure membranes like nanofiltration and reverse osmosis can remove a wide range of PFASs [16,30], but the disposal of highly contaminated eluates/concentrates produced during resin regeneration or treatments using high-pressure membranes remains a crucial issue. PFCAs and PFSAAs are both resistant to chlorination and ozonation [15-17*]. Furthermore, an increase in PFCA concentrations has been observed after ozonation, suggesting the transformation of PFASs containing C-H bonds (e.g. 6:2 FTAS or 6:2 FTAB) into PFCAs [16,17*,26,31].

Most studies have focused on PFCA and PFSA removal, whereas other PFASs can be present in drinking water sources impacted by point sources. The preliminary data available suggest that the efficient removal of new PFASs can be challenging for conventional drinking water treatments. GAC systems are thus inadequate for the efficient removal of 13 recently discovered PFASs [32]. Negligible removal by a treatment process including coagulation, ozonation, biofiltration and disinfection was observed for PFPrOPrA (GenX) and other perfluoroalkyl mono- and multi-ether carboxylic acids [17*]. In the Netherlands, the drinking water treatment plants investigated were not able to completely remove PFPrOPrA or 2 polyfluoroalkyl carboxylic acids [18*]. In a batch experiment, PAC achieved > 50% removal of 2 perfluoroalkyl multi-ether carboxylic acids, but < 40% for other perfluoroalkyl ether carboxylic acids [17*] tested.

Current regulations and guidance values

The discovery of many different PFASs in raw and drinking water has led several European countries and the U.S. to establish regulatory advisory levels or guidelines [33] that could not be exhaustively cited in this review. Since toxicological data on many PFASs are sparse, most of these provincial and national authorities set regulatory guidelines for perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS). Thus, in 2016, the U.S. Environmental Protection Agency established a lifetime health advisory level at 70 ng/L for the sum of PFOA and PFOS in drinking water. The German Drinking Water Commission recently revised its risk assessment for PFASs, including short-chained PFASs [34].

The Danish Environmental Protection Agency and the National Food Agency of Sweden had an innovative and pragmatic approach in establishing guidance values for the sum of several PFASs frequently found in drinking water. Above these values, PFAS concentrations could pose a health risk and should therefore be reduced and consumers must be advised not to drink the water until these concentrations have been reduced. In Sweden, for instance, a drinking water action level of 90 ng/L has been established for the sum of 3 PFSAs, 7 PFCAs and 6:2 FTSA [33].

In Sweden [13,33], France [10], Germany [6], Italy [35], and the U.S. [14], drinking water sources have been closed down or advanced treatments implemented to lower PFAS concentrations in tap water, either because the guidance values were exceeded or because the precautionary principle was applied in the absence of toxicological knowledge on the PFASs detected.

Effects of exposure to contaminated drinking water

Human exposure to PFASs can occur through contact with consumer products, ingestion or inhalation of house dust particles, or drinking water. However, food—especially seafood and freshwater fish—seems to be the dominant exposure source for the general population [36]. Risk assessment is complicated because the exposure involves many different PFASs, and even more complicated in the case of water tainted by emerging PFASs such as GenX or 6:2 FTAB [37]. Often, only direct exposure to perfluoroalkyl acids (PFAAs), i.e. PFCAs and PFSAs, is taken into account. The uncertainty on the contribution of each of the multiple exposure sources may explain why different PFAA concentrations have been proposed as a limit above which the relative importance of drinking water in overall exposure may be considered significant. Contaminated water can indirectly contribute to human exposure (Fig. 1). Thus, in areas with a history of PFAS-contaminated drinking water, it has been shown that short-chained PFASs can be readily transferred to vegetables via watering [6,38] (Fig. 1).

After drinking tainted water, the human serum levels of some PFAAs in the exposed population have been found to be above those of the general population [13,14,19,20,33,39,40]. A probable link between these higher serum levels of PFAAs and adverse health effects has been mentioned in a series of studies investigating the health effects of PFOA exposure among residents living in six contaminated water districts (the average PFOA concentration was 3.5 µg/L in the most contaminated water district) [41].

Conclusions

Due to their widespread use since the 1950s, their release into the environment and extreme persistency, PFASs have been frequently detected in the raw water of many countries. Conventional drinking water treatments are unable to entirely remove them. In the vicinity of sites heavily impacted by PFAS releases, sometimes remediation simply entailed closing down the water source, with important socioeconomic consequences. Furthermore, the per- and polyfluorinated alternatives to regulated PFASs developed by manufacturers have also recently been detected in water. These novel PFASs are also persistent and recalcitrant to removal from water by conventional treatments, so further research is needed to develop effective water treatments. It is difficult to include novel PFASs in drinking water guidelines since toxicological data for most of these chemicals are still inadequate. At this point in time, regulations based on the precautionary principle and/or chemical properties such as persistency and mobility are the only way to reduce the use and release of such chemicals. It is quite likely that new PFASs will be discovered and detected in the next few years. Although the main PFAS point sources (e.g. manufacturing facilities and firefighter training areas) have now been located and surveyed, this is not the case for landfills where industrial waste containing PFASs was dumped from the '60s to the '80s, a period when waste disposal requirements were not as stringent as now. It is likely that some of these sites will be found out by chance during routine monitoring. It can be challenging to accurately locate a point source, as many other different sources (e.g. biosolid applications, or the textile, paper, electronic and semiconductor industries) can release PFASs into the environment. PFAS contamination of water resources is often irreversible, and condemns important water supplies for many years. Like in other areas (such as the fight against doping in sport), scientific knowledge and regulations do not progress at the same pace as the market.

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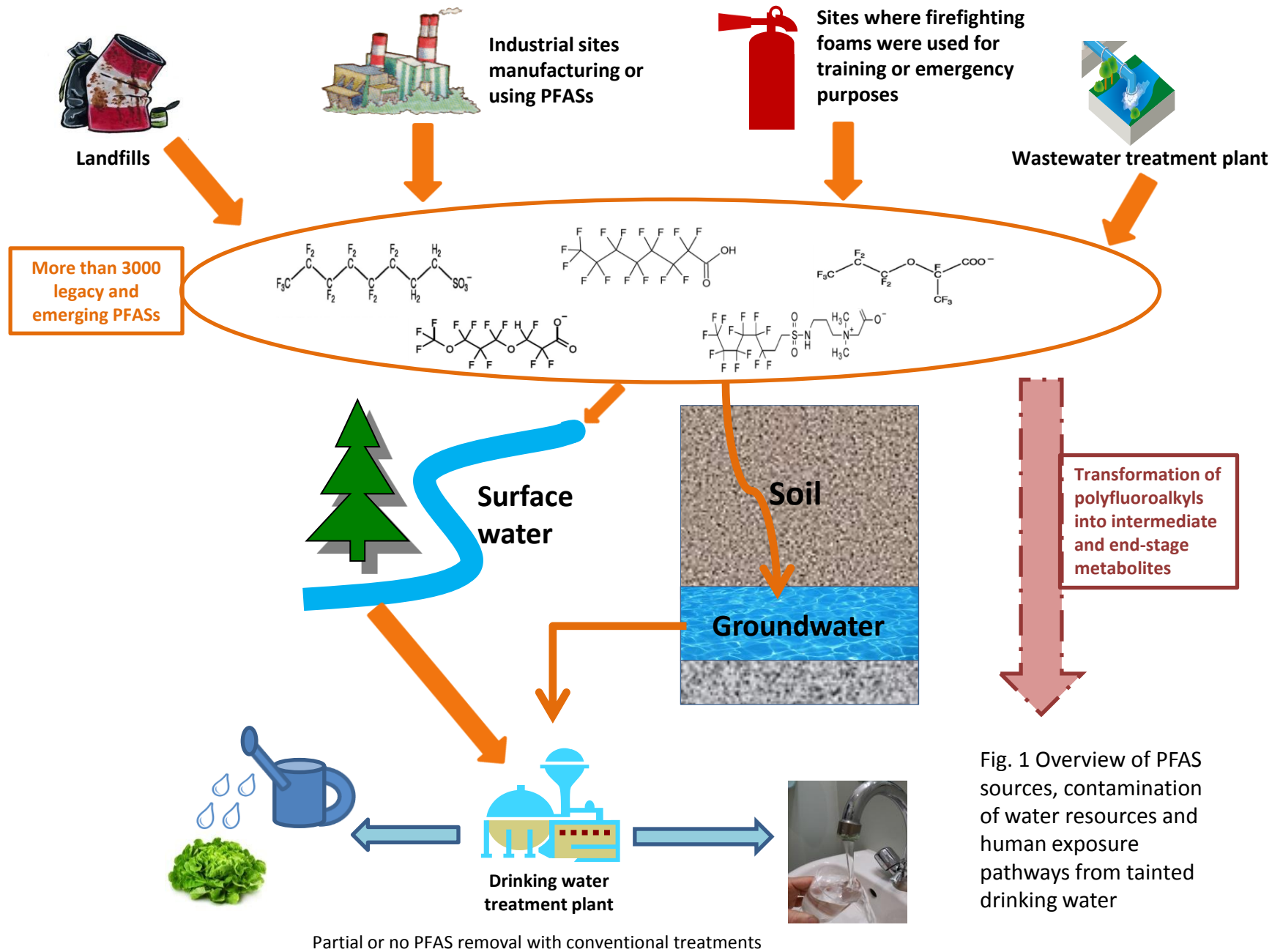


Fig. 1 Overview of PFAS sources, contamination of water resources and human exposure pathways from tainted drinking water