

PFAS in the Environment

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The current article reviews the state of art of the perfluoroalkyl and polyfluoroalkyl substances (PFASs) compounds and provides an overview of PFASs occurrence in the environment, wildlife, and humans. This study reviews the issues concerning PFASs exposure and potential risks generated with a focus on PFAS occurrence and transformation in various media, discusses their physicochemical characterization and treatment technologies, before discussing the potential human exposure routes. The various toxicological impacts to human health are also discussed. The article pays particular attention to the complexity and challenging issue of regulating PFAS compounds due to the arising uncertainty and lack of epidemiological evidence encountered. The variation in PFAS regulatory values across the globe can be easily addressed due to the influence of multiple scientific, technical, and social factors. The varied toxicology and the insufficient definition of PFAS exposure rate are among the main factors contributing to this discrepancy. The lack of proven standard approaches for examining PFAS in surface water, groundwater, wastewater, or solids adds more technical complexity. Although it is agreed that PFASs pose potential health risks in various media, the link between the extent of PFAS exposure and the significance of PFAS risk remain among the evolving research areas. There is a growing need to address the correlation between the frequency and the likelihood of human exposure to PFAS and the possible health risks encountered. The current entry highlighted the significance of the future research required to fill in the knowledge gap in PFAS toxicology and to better understand this through real field data and long-term monitoring programs.

poly-fluoroalkyl substances (PFASs)

toxicology

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regulatory values 1. Introduction

1. Introduction

Widespread surface and groundwater contamination with perfluoroalkyl and polyfluoroalkyl substances (PFASs) has become of great concern in the last few years. PFAS was first realized in the globe through the identification of perfluorooctane sulfonic acid, $C_8F_{17}SO_3H$ (PFOS), in wildlife [1][2]. PFASs have recently received increasing global attention because of their persistence and toxicity in the environment, bioaccumulation potential, and possible adverse health impacts [3]. PFAS are commonly have an aliphatic carbon composition in which hydrogen molecules have been replaced by fluorine completely (prefix: per-) or partially (prefix: poly-) [4]. These compounds are characterized by their highly polar and strong carbon fluorine bonds [5]. They are considered as highly fluorinated surfactants that have been applied in numerous industrial applications and manufactured goods including food packaging, firefighting foams, clothes and protective coatings for fabrics and carpets, electronics and fluoropolymer manufacturing [1][2][5][6][7][8][9]. The most extensively produced and frequently detected PFASs in the environments are perfluorooctanoic acid, $C_7F_{15}COOH$ (PFOA) and perfluorooctane sulfonic acid, $C_8F_{17}SO_3H$ (PFOS) [1].

PFASs have been discovered in different environmental compartments, including water, sediment organisms, and air [6][10][11][12].

PFAS has been a serious concern to industry, governments scientists, and even to the public worldwide [13]. It has been detected in various aquatic matrixes, including rain, snow, groundwater, tap water, lakes, and rivers with the C8-based substances PFOS and PFOA typically being the dominating compounds [4][9][14]. PFAS degradation products can be freely mobile in water, soil, and air, and can be extremely resistant to breakdown by different processes. The complexity of measuring PFAS in various media, and the associated unknown risks are among the challenges facing the current regulatory bodies [4]. Typical concentrations of PFASs in water are very low, however, higher concentrations of (mg/L) have been observed in surface and groundwater after firefighting activities closed to fluorochemical manufacturing facilities. PFASs spread worldwide has triggered the governmental concern towards regulating the exposure and spread of PFASs [15][16]. Although there is enough evidence about the negative impacts of PFAS on human and animal health, the scale of the risk imposed by PFAS compounds is not fully understood. The current regulations tend to address the potential risk limit for various wildlife where the PFASs persistence, bioaccumulation potential, and toxicity (PBT) raise a great concern [6]. Several studies have reviewed various aspects related to PFASs fate and behavior in different environments. They also reviewed the sources and occurrence of PFOA in drinking water, toxicokinetic, and health impacts [17][18][19][20][21]. Other reviews on PFASs have discussed different aspects such as environmental biodegradation of PFASs, PFASs removal from drinking water treatment plants, wastewater treatment plants and PFASs transformation in landfills [22][23][24]. The authors are aware of the developing research concerning PFAS and the many reviews investigating the PFAS human exposure, fate, transport, accumulation, health hazard and guidelines [2][5][16][25][26][27][28][29][30][31]. The current mini review investigates the PFAS occurrence in collective all geo-environmental compartments and is the first to collate the various international PFAS standards in one article. The current study reviews existing publications in the field of PFAS and aims to: (i) summarize the recent publication in the field of PFAS and ensure easy access of the research on the occurrence and behavior of PFASs in various environments, (ii) to identify knowledge gaps in the PFAS field, particularly the discrepancies in the current prevailing legislation and practices across various countries, and (iii) to present the key future research directions to better address the PFAS issue.

2. PFASs Occurrence and Transformation

Due to the strong C-F bonds in PFASs, they are highly stable and PFASs are unlikely to degrade easily in the environmental matrices [32]. PFASs in the environment has been resulted from several sources. The sources of PFASs in groundwater, drinking water, and surface water could be categorized into (i) point as well as (ii) diffuse sources. Wastewater treatment plants are considered as the most common point sources of PFASs to surface water. Other forms of point sources have been found to have a high impact on surface water in the USA including industrial pollution from PFAS production sites. In addition, it was observed that high concentrations of PFASs can be existed in surface water closed to commercials and military places due to the usage of aqueous film forming foams (AFFF) that contains PFASs. Landfills are considered as important point sources for PFASs in groundwater that comprise PFAS polluted waste in China, and that they could cause a hazard for tap water pollution [33]. Also, in

Europe, landfills have not been supported to a significant degree in terms of their capacity for groundwater PFAS contamination. The following subsections illustrate the different occurrence of PFASs in the environments.

3.1. PFAS in Environment

2.1.1. PFASs in Water

The level of PFAS as well as their fate in water bodies have been investigated by many researchers as water represents one of the main pathways for human exposure to [34][35][36][37][38][39]. The consistent detection of PFAS compounds such as perfluoroalkane sulfonates (PFSAs) and perfluoroalkyl carboxylates (PFCAs) in tap water samples at various locations has raised the concern over their potential health risk [35]. Such detection has been reported in drinking water samples in Europe, China, Malaysia, Thailand, USA, Singapore, Vietnam, and Brazil [4][14][20][34][35][38][39][40][41][42][43][44][45][46]. Other compounds, including perfluoro hexanoic acid (PFHxA) [36], perfluorooctanesulfonamide (PFOSA) [47], and perfluorinated phosphonic acids (PFPAs) [48], are also among the commonly detected compounds in water samples. This relatively persistent level of PFAS exposure increases with the increased drinking water contamination events where it was estimated that the average human daily PFAS intake ranges from 0.17 to 0.21 ng/kg bodyweight/day for PFOS and PFOA, respectively [49]. The results by Gellrich et al. [49] revealed that short chain PFAS (<8 carbon atoms) were dominant in samples collected from tap water with a maximum level of 42.7 ng/L followed by mineral water and spring water [49]. One of the key aspects on PFAS level in drinking water is the difference in PFAS level in both treated and source water. An interesting finding by Lu et al. [35] indicated that PFAS concentration was higher in treated water compared with the source water which could be accounted by the potential contamination that may occur whilst treated water moving through the water network and the treatment plant facilities [35]. Moreover, literature showed that various and inconsistent pattern of PFAS compounds was found on many occasions. While PFOA was dominant PFAS compound in tap water samples tested from Shanghai, Beijing, and Nanjing, PFOS was the key PFAS compound in water samples collected from Shenzhen and Hong Kong, accounting for more than 50% of the total PFAS compounds.

Moreover, researchers found mysterious temporal and spatial patterns of PFOA and PFOS across the various events where a comparison of PFAS levels in tap water in various Chinese cities revealed that the PFAS level varied significantly from one city to another with the highest was reported in Shanghai [35]. Moreover, the inconsistent and varied PFAS level was also significant across various countries. Quinete et al. [40] found that, unlike the pattern and the level of PFOA and PFAS level in the USA and Japan tap water samples, PFOS level was higher than PFOA in tap water samples collected in China [40]. An average level of total PFCs of 130 ng/L was measured in tap water samples from Shanghai (China), and a much lower maximum PFCs level was identified in water samples from Toyama (Japan) (0.62 ng/L) [42]. An average of 2000 ng/L was identified in treated drinking water distribution system at the city of Oakdale, USA. This seems to be a very extreme level of PFAS where a health-based drinking water level of 0.04 g/L was assessed as a protective lifetime exposure through risk assessment exposure [12].

One of the main concerns about PFAS contamination is their persistence and bioaccumulation properties as well as the potential to travel with either water streams or sediments. Traditional treatment facilities seem unable to eliminate PFASs during normal treatment processes [50]. PFAS discharge into water bodies was also reported by Boiteux et al. [45] where river water proved to be impacted by the nearby fluorochemical manufacturing industry. This confirms that discharge of PFOA and PFOS are still detected in nearby industrial and manufacturing facilities. Results showed that river water and sediment samples as well as treated water samples at various stages from the main treatment plant have all showed various level of PFCA compounds coming from the manufacturing industry. Interestingly, PFCA was also detected at sediment samples at 62 km away from the source in almost 50% of the samples [45].

The occurrence of PFASs in surface water is frequently happening across many countries around the world [51]. Researchers have conducted several field studies for different types of surface water to investigate the occurrence and presence of PFASs [2][23][29][52][53]. A previous study was conducted to assess the level of PFASs from different locations in Gangs River, India. Results showed that around 15 types of PFASs were found in the water samples where the highest level detected was for PFHxA and PFBS. In addition, significant relationships were detected ($p < 0.05$) between the different PFASs substances such as PFCAs, PFSAs, PFBA, and PFHxS, indicating chemical binding and co-transport with dissolved oxygen carbon (DOC) in fresh and seawater. Consequently, assessed the pollutants concentration and spatial distribution of PFASs in Shuangtaizi Estuary, China. Results showed that the Shuangtaizi Estuary was in general polluted by PFASs. The total concentration of PFASs varied from 66.2 to 185 ng/L and from 44.8 to 209 ng/L in surface and bottom water of the Shuangtaizi Estuary, respectively, where the maximum concentration was reported for PFBS and PFBA. The level of PFASs in different environmental matrices was tested including surface runoff water rain, snow, and lake water in an urban area, to identify the sources of PFASs to urban water bodies [34][37][38]. Another research conducted by Yin et al. [54] discovered a significant temporal variation of PFASs compounds level over 12 months period due to the seasonal and climatic dry and wet conditions. Moreover, PFASs concentration was a function of the chain length where the level of short-chain compounds including PFBS, PFHxA and PFHpA tend to be highly influenced and decreased by the wet conditions. On contrary the level of long-chain PFASs compounds was more stable in both wet and dry conditions. These findings provide a good understanding to the leachate of PFAS compounds from point source pollution as landfills and treatment plant. The leaching of long chain is more likely controlled by the partitioning effect whilst the short chain leaching is influenced by the climatic conditions [54].

Another main finding in the field of PFAS in water is the variation of the PFAS where PFOA was the major compound with an average concentration of 35% of the total PFASs levels, in all environmental matrices investigated. In addition, the concentrations, and relative substances of PFASs in surface water were comparable to the concentrations found for urban lakes. Surface water leads to PFOA pollution in urban lakes. A sampling campaign was conducted in different seas in China in 2012. The results revealed that the higher concentration of PFAS was detected in the South Yellow Sea, where FTOH was the predominant substance, contributing 92–95% of the total PFAS [2].

2.1.2. PFASs in Soil

PFAS was detected in soil at various concentrations due to the reach out from various pollution sources where PFAS compounds retain in soil due to sorption, partition and other complex reaction [55]. [Table 1](#) shows the range of PFAS concentration in soil. The application and the reuse of sludge from wastewater treatment plants in farmlands is one of the main sources that contribute to soil contamination [56]. Other sources could be due to the degradation of fluorotelomer-based materials that lead to the release of PFCAs [41], precipitation, and water irrigation [57]. PFAS compound in soil in coastal areas can be emitted from direct sources which could level the PFAS concentration up to around 8–50 $\mu\text{g kg}^{-1}$ soil as was reported in Chinese soil [43] which is somehow higher than the proposed PFCs in soil proposed by the USEPA (6 mg/kg for PFOS and 16 mg/kg for PFOA). One main concern about PFAS in soil is the potential PFAS release and carryover by plants as well as the possible PFAS leaching to the underneath soil layers and the groundwater. This carryover of PFOA and PFOS to the plant was evident where the PFAS level in plants was proportionally related to the PFOA/PFOS in the soil [58]. PFCs uptake from contaminated soil by crops was reported [59], where samples from rye grass, grain, and potatoes showed high potential of PFCs transfer from soil to crops [59]. This resulted in proposing a preventative PFCs limit of 100 ng/g dry soil in sludge to be reuse for farming purposes as to limit the potential transfer of PFCs from soil to plants and crops [58]. Moreover, the potential leaching of PFAS from soil through vadose zone is another threat that requires more attention and understanding where insufficient data about in-situ soil remediation and contaminants leaching to the groundwater are available [59]. The development of PFAS compounds in the soil system is complex since PFAS compounds can attain both hydrophilic and hydrophobic characteristics [55]. While the transfer of PFAS from soil to plant roots undergo through diffusion and sorption onto roots, there are still insufficient details about the PFC transfer rates in various crops and vegetables [58]. This result was confirmed [58] where they found the straw and grains of maize plants have the same carboxylic and sulfonic functional groups as in the contaminated soil referring to a direct correlation between soil and crops PFASs contamination.

Table 1. Ranges of PFAS concentration in soil.

Header	Å. Høisæter, et al. [71] ng/g PFASs in Soil at Various Depths				Cai et al. [63] ng/g Dry Weight in	Chen et al. [69] ng/g Dry Weight	Cai et al. [61] ng/g Dry Weight	Gao et al. [72] ng/g Dry Weight from 32 Samples	Wang et al. [73] The Mean Values ng/g Dry Weight	Liu et al. [74] ng/g Dry Weight	Chen et al. [75] ng/g Dry Weight	Dalahmeh et al. [76] ng/g Dry Weight	Armstrong et al. [77]
0– 1.0 m	1–2 m	2–3 m	3–4 m	Dry soil in China	Tot.PFC 0.34–65.8				PFAS in soil at varying distances				
PFOS	500– 3000	1000– 6500	1000– 3500	1000– 1200	130	70.5	8.6–10.4	0.06	2583,	87	0–2	0.6–3	23
PFOA	NA	NA	NA	NA	NA	93	3.3–47.5	0.32	50	0.3–8	63	0.5–0.9	24
PFHxS	NA	NA	NA	-	NA	61	NA	0.19	36	65			
Σ PFCs	NA	NA	NA	NA	NA	-	NA	-	NA	99	NA	8	126–809
PFHxA	NA	NA	NA	NA	NA	NA	NA	0.09	NA	NA	NA	0.2–0.5	8
PFAA	NA	NA	NA	NA	NA	NA	NA	1.30–913	NA	NA	NA	NA	NA
PFBS	NA	NA	NA	NA	NA	NA	NA	0.05	NA	NA	NA	NA	NA

PFASs toxicity their impact on soil microorganisms is among the other factors that can deteriorate the soil quality. Research found that the PFCs can negatively affect the soil functionality where it may disturb soil enzyme activity as well as change the microbial availability and damage the cellular structure [60][61][62]. The same result was

confirmed by Sun et al. [63] as the soil contaminated with PFASs compounds had less bacterial diversity [63]. PFOA and PFOS are the dominating compounds reported in soil where their concentration ranged from <1 to around 13,000 ng/g in soil [55]. The fate of PFASs in the soil is a function of many parameters including soil pH, soil structure, clay content, organic matter content (OM), PFAS characteristics (long versus short chain), and climatic conditions [60]. OM seems to be the most significant controlling factor determining the PFAS toxicity level where the PFAS toxicity is inversely proportional with the soil OM content [32][64]. Additional research on PFAS adsorption and migration from soil to the groundwater and how this can migrate with the groundwater is still a research gap needs more investigation and modelling to account for the various PFAS concentrations in various groundwater conditions [59]. Contaminated soil with PFAS is a challenge since there is no definite remediation strategy to address the in situ PFAS remediation. Although soil stabilization using various reagents such as clay and Portland cement seems to be a promising technique for soil remediation, it does not provide an elimination for PFAS where it does not remove PFAS permanently [59]. Finally, the PFAS uptake by plant poses a direct human risk where the food chain represents a main risk pathway. Therefore, a toxicological risk assessment addressing the maximum allowed levels of 1.5 and 0.15 µg/kg body weight as TDI µg/kg for PFOS and PFOA, respectively, were identified by the European Food Safety Agency (EFSA) as a function of the respective tolerable daily intakes (TDI) of the compounds [58][65].

PFASs compounds are soluble in water and have the potential to leach down to the groundwater particularly in areas with potential source pollution like landfills and treatment plants. PFASs occurrence and leaching was reported by many researchers around the world [22][54][66][67][68]. The potential PFAS leaching could be alarming in many cases where PFASs were detected at large depths (15 m) below ground [69]. Yet, the leaching speed and behavior vary from one PFASs to another which depends on the soil binding, retardation and adsorption capacity [55]. The leaching characteristics of PFASs compound is a function of the chain length where short chain is more mobile than long ones. An analysis of the landfill leachate from 27 landfills in Australia was investigated by Gallen et al. [22]. Interesting findings presented in their study showed that the landfill leachate was significantly different from one landfill to another with an average PFASs of 1700 ng/L and a maximum PFAS level of 25,000 ng/L [22]. In contrast, the reported PFAO range in USA landfills was ranging between (7280–290,000) ng/L compared with 214,000 ng/L in China [34]. Nonetheless, these PFAS concentration are highly likely to vary due to the heterogeneous nature of waste dumped in landfills as well as the varied PFAS content in the generated landfilled materials. Operating landfills receiving municipal waste had much more PFAS level than closed ones and the leachate from landfills with construction and demolished materials seems to leach more PFAS than municipal landfills. Another study investigated the leachate from 11 landfills in USA and found that PFAO was detected in all samples [69]. [Table 2](#) presents the level of various PFCS and PFAS compounds in leachate and compare the PFASs in water and solid. It can be seen that PFASs levels vary from one compound to another as a function of chain length and climatic conditions as illustrated in the previous sections. The risk associated from the landfill leachate is the potential volumes leachate generated particularly in wet climates, which contributes to the groundwater contamination. The total leachate volume in the USA was estimated to be around 61 million m³ with around 80% coming from landfills [69]. Meanwhile, the leachate mass of Σ PFA in China was estimated by around 3 ton per year with the landfill leachate contribute to around 35% of this quantity [33]. Interestingly, analysis of leachate from young landfills showed much higher PFAS concentration in many occasions confirming the fact that

the complexity and persistence of PFAS compound has been developed in the recent years where more frequent PFAS containing materials are in use [66][69]. The uniqueness of the landfills associated with its design capacity, climate, age, engineering, dumped materials and frequency and other factors made it hard to predict the amount of PFAS leachate in various landfills where ad-hoc studies to be conducted. The results from various areas across the globe showed significant variation of PFAS leachate from one country to another where a maximum was reported in Australia (25,000 ng/L). This was evident while the leachate was significantly lower in Norway (590 to 757 ng/L), Germany (<0.37 to 2509 ng/L), and China (146 to 4430 ng/L) [33]. In conclusion, although the phasing out of PFAS materials and the ongoing effort to eliminate the PFAS release in the environment, yet there seems to be a need to consider more adaptation strategies dealing with PFAS risk. The increasing evidence of PFAS in newly designed and operated landfills indicates the potential exposure to higher leaching risk with greater PFAS concentrations is leaching to the environment is growing [69].

Table 2. Ranges and mean concentration of individual PFCs in landfill leachate.

Header	Gallen et al. [22] (ng L ⁻¹)	Busch et al. [67] (ng L ⁻¹)	Herzke et al. [70] (ng L ⁻¹)	Yin et al. [64] (ng L ⁻¹)	Benskin et al. [72] (ng L ⁻¹)	Eggen et al. [73] (ng L ⁻¹)	Robey et al. [68] (ng L ⁻¹)	Fuertes et al. [74] (ng L ⁻¹)	Eggen et al. [72] (ng L ⁻¹)	Huset et al. [75] (ng L ⁻¹)	Garg et al. [76] (ng L ⁻¹)				
landfill (>50% SW)	landfill (>50% C&D)	Compounds in landfill leachates	Coated textiles, Teflon waste, fire-fighting foam, papers, and furniture	Leachate from CW outlet system (Max. level)	Municipal landfill leachate	Municipal land fill leachates	Foam produced via the bubble aeration of landfill leachate	Raw Leachate in MSW landfill	Treated Leachate in MSW landfill	PFCS analysis-untreated leachate Water	PFCS analysis -untreated leachate Particles	Leachates from six landfill	Manufacture and disposal of electric and electronic products		
PFOS	300	1100	235	570	187	439	4400	2920	104	25	NA	2920	34	56–160	128,670
PFOA	510	1200	926	9500	516	3457	1500	767	951	590	520	767	4	380–1100	118.3
PFHxS	940	3700	178	-	143	308	190	281	2058	630	870	281	ND*	120–700	133,330
PFDS	-	-	-	NA	0.72	63	<14	ND	-	-	-	-	-	0–16	-
PFHxA	1300	5000	2509	-	697	868	2500	757	2178	65	77	757	ND	270–2200	76
PFHpA	360	760	280	NA	486	690	277	454	-	-	277	ND	100–2800	9	
PFNA	29	98	80	-	62	100	450	539	64	-	-	539	ND	19–140	8
PFDA	22	46	51	-	NA*	27	1100	75	87	-	-	70	ND	0.3–64	8
PFAA	NA	NA	NA	-	-	55	-	-	-	-	-	-	-	-	-

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Header	Gallen et al. [22] (ng L ⁻¹)	Busch et al. [27] (ng L ⁻¹)	Herzke et al. [70] (ng L ⁻¹)	Clarke [21] (ng L ⁻¹)	Yin et al. [24] (ng L ⁻¹)	Benskin et al. [22] (ng L ⁻¹)	Eggen et al. [23] (ng L ⁻¹)	Robey et al. [68] (ng L ⁻¹)	Fuertes et al. [74] (ng L ⁻¹)	Eggen et al. [23] (ng L ⁻¹)	Huset et al. [25] (ng L ⁻¹)	Garg et al. [76] (ng L ⁻¹)
PFBS	1350	NA	112	1916	190	-	-	-	-	<5	ND	280–2300
PFPeA						11	325					

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PFDA perfluorodecanoic acid, PFBS nonafluorobutane-1-sulfonic acid, PFHpA perfluoroheptanoic acid, PFHxS perfluorohexane sulfonate, PFOA perfluorooctanoic acid; ND: Not detected; NA: Not analysed; Guideline levels for PFOA and PFOS in drinking water: The role of scientific uncertainty, risk assessment decisions, and social factors. *J. Expo. Sci. Environ. Epidemiol.* 2019, 29, 157–171.

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