Per- and Poly-Fluoroalkyl Substances' Pollution

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Per- and poly-fluoroalkyl substances (PFASs) refer to a class of compounds in which all or part of the hydrogen atoms linked to carbon atoms in alkane molecules are replaced by fluorine atoms. Because of the strong polarity of the C-F bond, PFASs have more stable and more excellent properties than other hydrocarbons (such as a remarkably high chemical stability and excellent hydrophobicity and oleophobicity), so they have been used in various fields of production, such as plastic wrap, paper, coatings, poly-tetrafluoroethylene products and foam fire-extinguishing agents.

Keywords: i-PFASs ; China ; river ; lake ; precipitation ; the Tibetan Plateau

1. Introduction

Per- and poly-fluoroalkyl substances (PFASs) refer to a class of compounds in which all or part of the hydrogen atoms linked to carbon atoms in alkane molecules are replaced by fluorine atoms [1]. Because of the strong polarity of the C-F bond, PFASs have more stable and more excellent properties than other hydrocarbons (such as a remarkably high chemical stability and excellent hydrophobicity and oleophobicity), so they have been used in various fields of production, such as plastic wrap, paper, coatings, poly-tetrafluoroethylene products and foam fire-extinguishing agents [2]. According to different functional groups and physicochemical properties, PFASs can be divided into (1) ionic PFASs (i-PFASs), such as per-fluoroalkyl carboxylic acids (PFCAs) and per-fluoroalkane sulfonic acids (PFSAs), and (2) neutral PFASs (n-PFASs), such as fluorotelomer alcohols (FTOHs) and per-fluoro-octane sulfo-namidoethanols (FOSEs) [3]. The PFASs referred to in this paper are i-PFASs. Due to the mass production and use of some PFASs, PFASs have been widely detected in various environmental media, animals and plants in recent years [4][5][6], and have also been found in the human body [7][8]. With intensive studies, the persistence, bioaccumulation, long-distance transportation and biohazard of some long-chain PFASs (long-chain PFASs refer to PFCAs with seven or more perfluorinated carbons and PFSAs with six or more perfluorinated carbons) have been gradually confirmed [9]. Subsequently, some countries and organizations have successively issued series of rules and regulations to restrict the use of such substances [10], and PFOS, PFOA and their salts were listed under Annex B and Annex A of the Stockholm Convention on Persistent Organic Pollutants in 2009 and 2019, respectively [11][12]. Meanwhile, in 2019, the Persistent Organic Pollutants Review Committee (POPRC) recommended that per-fluorohexane sulfonate acid (PFHxS) and its salts be listed in Annex A of the Convention [13]. In addition, at the 17th meeting of the POPRC, held in January 2022, it was suggested that long-chain PFCA (involving carbon-chain lengths from 9 to 21), its salts and related compounds should be listed in Annexes A, B and/or C of the Stockholm Convention on Persistent Organic Pollutants [14]. With the restricted use of PFOS, PFOA and other substances, some short-chain per-fluoroalkyl acids and new polyfluoride substitutes [such as 6:2 chlorinated polyfluorinated ether sulphonic acid (F-53B)] are becoming research focuses [15][16]. The international community is paying increasing attention to PFASs.

Since 2000, some major manufacturers have phased out the production of PFOS, PFOA and their salts [17]. The production of these substances has gradually shifted from developed countries, such as North America and Europe, to developing countries, especially China [18]. China has become the largest manufacturer and supplier of PFOS and PFOA in the world since 2004 [3]. Some studies have estimated the emissions of PFOA and its salts in China from 2004 to 2012. The results show that the cumulative emissions reached 250 tons, and China became the largest PFOA emission site at that time [19]. Therefore, the environmental monitoring of and risk research on PFASs in China have attracted international attention. Studies have shown that PFASs can exist stably in water environments [20], and the ocean is the final sink for PFASs [21]. Therefore, it is necessary to explore the pollution characteristics and possible sources of PFASs in China's waters.

2. PFASs in Surface Water

This study summarized the concentration data from monitoring studies of PFASs in surface water in China over the past 10 years, as shown in **Figure 1** and **Table 1** for details. It was revealed that the concentrations of PFASs in surface water varied greatly in different areas in China (from 0.775 to 1.06 × 10^6 ng/L). The average concentrations in the Daling River, tested in 2018, and Xiaoqing River, tested in 2013, were highest (2.31 × 10^3 ng/L and 2.14 × 10^3 ng/L, respectively) [22][23], followed by that in the Daling River in 2011 (1.04 × 10^3 ng/L) [24]. The concentrations in other rivers were relatively low. It was found that the concentrations and composition characteristics of PFASs were mainly related to the emissions of fluorinated manufacturing facilities (FMFs), wastewater treatment plants (WWTPs) and urbanization development. The concentrations of PFASs in the water bodies after they flowed through FMFs were significantly higher than those before they flowed through FMFs, so the discharge from the FMF production process was one of the main sources of PFASs in surface water. For example, the concentration of PFASs in the Daling River flowing through the Fluorochemical Industrial Zone in Fuxin City was significantly higher than the concentration in other watersheds by an order of magnitude. With the development of the fluorochemical industry, the PFASs from manufacturing processes caused more pollution to enter the Daling River, and the concentrations detected in 2018 increased by 50% compared to 2011 [23][24]. Liaohe River was also affected by the Fluorochemical Industrial Zone in Fuxin City with the maximum concentration of 781 ng/L [25]. Similarly, Xiaoqing River was influenced by FMFs, such as metallurgy, electronics, and firefighting, with the maximum concentration of 1.06 × 10^6 ng/L detected in 2013 [22]. The north of Taihu Lake was close to FMFs, such as the manufacturing of paint and plastic products, with concentrations of 56.1–120 ng/L, while concentrations in other parts of the study area ranged from 10.0 to 79.4 ng/L [26]. The concentration in Guanlan River during the abundant water period was six times higher than that in the dry water period, probably due to the large amount of surface runoff and rainwater flushing during the abundant water period that carried pollutants from the periphery of the industrial area into the water body [27]. East China was also a concentrated area for FMFs, and the PFAS concentration in Huangpu River flowing through this region increased (from about 300 to 380 ng/L) [28]. Moreover, the variation tendencies of PFAS concentrations in different water bodies were also different; for example, the concentration of PFASs in the Daling River increased from 2011 (average of 1.04 × 10^3 ng/L [24]) to 2018 (average of 2.31 × 10^3 ng/L [23]), and that in Xiaoqing River decreased from 2013 (average of 2.14 × 10^3 ng/L) [22] to 2014 (average of 457 ng/L) [29].

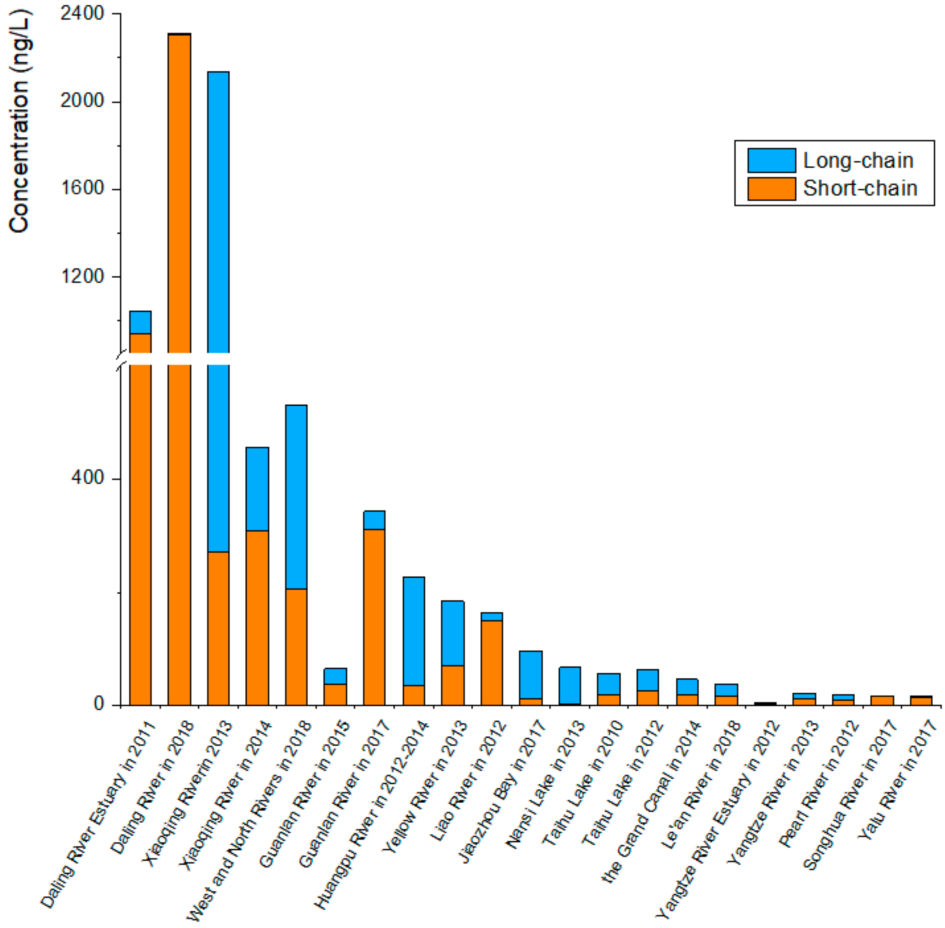

Figure 1. Levels and compositions in surface water in China in past ten years.

Table 1. PFAS concentrations in surface water in China in past ten years.

Sampling Year	Area	Analytes	Concentration (ng/L)	Reference
2010	Caohai (north of Dianchi Lake)	C4–C12 PFCAs, C8 PFSAs	35.8–135.9	[30]
2010	Taihu Lake	C4–C12 PFCAs, C6, C8 PFSAs	10.0–119.8	[26]
2011	Daling River estuary	C4–C10 PFCAs, C4, C6, C8 PFSAs	5.26–4.74 × 10^3	[24]
2011	Tangxun Lake	C4–C13 PFCAs, C4, C6, C8 PFSAs	4570–11,890	[31]
2012	Liao River	C6–C12 PFCAs, C4, C6, C8 PFSAs and FOSA	44.4–781	[25]
2012	Pearl River	C4–C11 PFCAs, C4, C6–C8, C10 PFSAs	3.0–52	[32]
2012	Taihu Lake	C6–C12 PFCAs, C4, C6, C8 PFSAs and FOSA	17.2–94.3	[25]
2012	Yangtze River estuary	C4–C10 PFCAs, C4, C6, C8 PFSAs and FOSA	1.7–12	[33]
2012–2014	Huangpu River	C3–C12, C14 PFCAs, C4, C6, C8 PFSAs	39.8–596.2	[28]
2013	Nansi Lake	C5, C7–C11 PFCAs, C4, C6, C8 PFSAs	38.4–91.4	[34]
2013	Pearl River delta	C5–C14, C16, C18 PFCAs, C4, C6, C8, C10 PFSAs	1.53–33.5	[35]
2013	Xiaoqing River	C4–C11 PFCAs, C4, C6–C8, C10 PFSAs	32.2–1.06 × 10^6	[22]
2013	Yangtze River	C4–C11 PFCAs, C4, C6, C8, C10 PFSAs and FOSA	2.2–74.56	[36]
2013	Yellow River	C4–C12 PFCAs, C4, C8 PFSAs	44.7–1.52 × 10^3	[37]
2014	Xiaoqing River	C4–C12 PFCAs, C8 PFSAs	36.5–4.96 × 10^5	[29]
2014	Grand Canal	C4–C11, C14 PFCAs, C4, C6, C8 PFSAs	7.8–218	[38]
2014–2015	Jiulong River estuary	C4–C14 PFCAs, C4, C6, C8, C10 PFSAs	3.30–110	[39]
2015	Guanlan River	C6–C12, C14 PFCAs, C4, C6, C8 PFSAs	37.04–103.7	[27]
2016	Sanggan River	C4–C12 PFCAs, C4, C6, C8 PFSAs	6.67–9.74	[40]
2016	Yanghe River	C4–C12 PFCAs, C4, C6, C8 PFSAs	2.10–197	[40]
2016	Yongding River	C4–C12 PFCAs, C4, C6, C8 PFSAs	12.4–108	[40]
2017	Guanlan River	C6–C12, C14 PFCAs, C4, C6, C8 PFSAs	179.15–613.68	[27]
2017	Jiaozhou Bay	C4–C12 PFCAs, C4, C6, C8, C10 PFSAs and FOSA	35.00–205.34	[41]
2017	Poyang Lake	C4–C11 PFCAs, C4, C6, C8 PFSAs	12.9–56.2	[42]
2017	Songhua River	C4–C14 PFCAs, C4, C6, C8, C10 PFSAs	6.4–32	[43]
2017	Yalu River	C4–C14 PFCAs, C4, C6, C8, C10 PFSAs	6.3–28	[43]
2017	Yangtze River	C4–C11 PFCAs, C4, C6, C8 PFSAs	7.8–586.2	[42]
2018	Daling River	C4–C13 PFCAs, C4, C6, C8, C10 PFSAs, HFPO-DA and 6:2 Cl-PFESA	48.4–4.58 × 10^3	[23]
2018	Le’an River	C4–C14 PFCAs, C4, C6, C8, C10 PFSAs	14.71–114.72	[44]
2018	West and North Rivers	C4, C6–C10 PFCAs, C4, C6, C8, PFSAs, 6:2 Cl-PFESA and FHUEA	0.775–1.06 × 10^3	[45]

In addition, the concentrations of PFASs in the water bodies after flowing through WWTPs were obviously higher than those before the water flowed through WWTPs, so the sewage discharge of WWTPs was another main source of PFASs in surface water. For example, after flowing through WWTPs, the concentration of PFASs in Huangpu River, mentioned above, rose from ~200 to ~250 ng/L [28]. The peak concentration of the Yongding River occurred at 108 ng/L downstream of the WWTP in its watershed and 12.4 ng/L upstream [40]. Due to the sewage discharged by WWTPs entering into the north of Dianchi Lake directly, concentrations in the north (35.8–135.9 ng/L) were higher than those in the south (less than 25 ng/L) [30]. It was found that the concentrations of PFASs in water flowing through cities with high degrees of

urbanization had significant rises because of traffic, commercial activities and so on. For example, the West and North Rivers flow through the cities of Shaoguan, Heze and Qingyuan, and rapid urban development had a remarkable impact on the PFASs in the water bodies with the concentrations up to 1.06×10^3 ng/L [45]. Yanghe River (one of the tributaries of the Yongding River) ran through the highly urbanized city of Zhangjiakou with a high concentration of 197 ng/L, and Sangan River, another tributary of the Yongding River, ran through less densely populated and less urbanized areas with relatively smaller concentrations ranging from 6.67 to 9.74 ng/L [40]. Likewise, the Songhua River and the Yalu River, which were relatively less densely populated, also had relatively low concentrations of PFASs, with a maximum of only 32 ng/L [43].

In the past, long-chain PFASs made the greatest contributions, especially PFOA and PFOS. In recent years, short-chain PFASs have gradually become the main compounds in water pollution, such as PFBA, PFBS, PFHxA and PFPeA. For instance, in the Daling River, the proportions of PFBS (35.9% in 2011 and 48% in 2018) and PFBA (32.8% in 2011 and 41% in 2018) have gradually increased [23][24]. On the contrary, PFOA (17.5% in 2011 and 2.3% in 2018) showed a decreasing trend [23][24]. In the main stem of the Pearl River, PFBS contributed 36% while PFOS contributed 21% [32], and the same phenomenon was found in the tributaries of the Pearl River (52% PFBA and 25% PFOA) [45]. Similarly, PFHxA accounted for 46% in spring and 53% in summer for PFPeA, with short-chain PFASs predominating in the Jiulongjiang estuary [39].

3. PFASs in Precipitation

The monitoring results of studies of PFASs in precipitation in China over the past 10 years were summarized as shown in **Table 2**. The monitored concentrations of PFASs in precipitation from the available studies in China, except for TFA (trifluoroacetic acid), ranged from 4.2 to 191 ng/L, which were higher than those in precipitation from developed countries, such as Germany (1.6–48.6 ng/L) [46], Japan (8.16–37.2 ng/L) [47] and France (2.59–3.76 ng/L) [48] during the same period, but lower than the concentrations in the United States (50–850 ng/L) [49].

Table 2. PFAS concentrations in precipitation in past ten years.

Sampling Year	Region	Analytes	Concentration (ng/L)	Reference
2010	Tianjin	C4–C12 PFCAs, C4, C6, C8, C10 PFSAs, 6:2 FTUCA and 8:2 FTUCA	22.5–147	[50]
2016	28 cities in mainland	C2 PFCA (TFA)	$8.80–1.8 \times 10^3$	[51]
		C3–C12 PFCAs, C4, C6, C8 PFSAs, 8:2 FTUCA, 6:2 FTSA, 6:2 Cl-PFESA, 6:2 diPAP and 8:2 diPAP	5.37–191	
2017	Jiaozhou Bay	C4–C12 PFCAs, C4, C6, C8, C10 PFSAs and FOSA	4.20–66.1	[41]

The PFAS concentrations in precipitation were generally lower than those in surface water in the same area. The average concentration in precipitation in Jiaozhou Bay was 22.0 ng/L [41], which was lower than that in surface water (60.5 ng/L) [41]. In the Nanchang section of Yangtze River, the average PFAS concentration in precipitation was 9.2 ng/L [51], which was lower than that in surface water (44.6 ng/L) [42]. Similarly, in the Xiamen section of Jiulong River, the average precipitation concentration of PFASs (22.0 ng/L) [51] was lower than that of surface water (38.2 ng/L) [39].

PFAS precipitation concentrations tended to be higher in areas with concentrated distribution of FMFs. High PFAS concentrations (80.6 ng/L) were found in urban areas in Northeast China (such as Fuxin, Dalian and Harbin) with a high distribution of FMFs [51]. Similarly, the PFAS precipitation concentration in Tianjin was also high (63.1 ng/L) [50]. Moreover, higher precipitation concentrations were found in inland and coastal cities with higher urbanization levels. The average concentration was 84.8 ng/L in more urbanized inland cities, such as Zhengzhou and Chengdu, with high traffic volumes and commercial activities [51]. In coastal cities, such as Shantou, Xiamen and Weifang, the average concentration of PFASs in precipitation reached 182 ng/L [50]. Ship maintenance in the port contributed greatly to PFAS pollution.

A precipitation monitoring study conducted by Chen et al. [51] in 28 cities in China found that TFA contributed the largest proportion, accounting for 78.1%, followed by PFOA, PFBA and PFOS, accounting for 5.08%, 3.16% and 3.12%, respectively. The proportions of short-chain PFASs (C4–C7) and long-chain PFASs (≥C8) were almost the same [51]. Han et al. [41] found that PFOA occupied 61.2% of the precipitation monitored in Jiaozhou Bay, followed by PFOS and PFHxA with 9.20% and 8.21%, respectively, and long-chain PFASs represented the largest proportion (87.3%).

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