

# Legacy and novel PFASs in wastewater, natural water, and drinking water: Occurrence in Western Countries vs China



Francesco Calore <sup>a</sup>, Pier Paolo Guolo <sup>a</sup>, Jiacheng Wu <sup>b</sup>, Qingfeng Xu <sup>b</sup>, Jianmei Lu <sup>b</sup>, Antonio Marcomini <sup>a,\*</sup>

<sup>a</sup> Department of Environmental Sciences, Informatics and Statistics, University Ca' Foscari of Venice, Via Torino 155, Venezia-Mestre, 30172, Italy

<sup>b</sup> College of Chemistry, Chemical Engineering and Materials Science, Soochow University, Suzhou, Jiangsu, 215123, China

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## ABSTRACT

A critical examination of the occurrence of PFASs in wastewater, surface water, groundwater, and drinking water is presented. Given the ongoing shift towards substitutes of legacy PFASs and the rising role of China as global leader in PFASs production, background and hotspot concentrations of PFASs in the aquatic environment of China and Western Countries (mostly the E.U., the U.S., and Canada) are reported and critically compared for the first time, with special attention to short-chain perfluoroalkyl acids, novel substitutes like perfluoroether carboxylic and sulphonic acids, and emerging PFASs like 6:2 FTAB. Data on PFASs concentrations in the examined matrices show that i) concentrations of total PFASs and of selected perfluoroalkyl acids (PFAAs) are generally higher in China, except for PFOS, and that ii) short-chain and novel PFASs are replacing their long-chain homologues but are rarely monitored. Recommendations are provided to help prioritize the PFASs to be investigated that can end up in the environment.

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## 1. Introduction

Per- and polyfluoroalkyl substances (PFASs) represent a class of thousands of xenobiotic contaminants that have been used for more than 60 years worldwide (i.e., since the end of the 1940s). They have been recently re-defined by the Organisation for Economic Co-operation and Development (OECD) as fluorinated substances with at least a fully fluorinated methyl or methylene carbon atom [1,2], but have been vastly studied on the basis of a previous definition according to which PFASs exhibit a partially (polyfluoroalkyl substances) or fully (perfluoroalkyl substances) fluorinated alkyl chain containing the  $-C_nF_{2n+1}$  moiety [3,4]. High chemical and thermal stability, amphiphilicity, and high surface activity are at the base of their extensive use as water- and stain-repellents and surfactants in numerous industrial sectors and commercial products (e.g., metal plating, textiles and leather treatment, packaging, aqueous film-forming foams, cosmetics,

medical devices) [5]. Because of the presence of perfluoroalkyl moieties they are overall persistent in the environment [6,7]. PFASs have been ubiquitously detected in water, soil, sediment, air, and biota (including humans) [8]. PFASs can bioaccumulate [9], and were linked to relevant health effects on humans, among which are endocrine disruption, neurotoxicity, hepatotoxicity, immunotoxicity, and cardiovascular diseases [10–13]. Recently, it has been hypothesized that the chemical pollution-related planetary boundary for at least four PFASs has been exceeded, especially as regards their virtually non-reversible global recirculation in the hydrosphere and, therefore, non-reversible global atmospheric deposition [14]. Since the beginning of the 2000's, several measures have been taken to phase out selected PFASs, to restrict their use in specific sectors, and set guidelines and thresholds to protect human and environmental health (see S2.2). For example, PFOA (perfluorooctane carboxylic acid) and PFOS (perfluoroalkyl sulfonic acid) have been identified as persistent organic pollutants (POPs) under the Stockholm Convention [15]. However, the gap between the number of PFASs introduced in the global market and the knowledge on their physical-chemical properties, toxicity and environmental occurrence widened, making management strategies for this class of substances significantly more difficult.

\* Corresponding author.

E-mail address: [marcom@unive.it](mailto:marcom@unive.it) (A. Marcomini).

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Experimental data on the physical-chemical properties (S2.1) and toxicity of PFASs is limited to a handful of perfluoroalkyl acids (PFAAs) [16,17], mostly long-chain perfluoroalkyl carboxylic and sulfonic acids (PFCAs and PFSAs), i.e. with a total number of carbons  $\geq 8$  and  $\geq 6$ , respectively [3], because of their higher bioaccumulation potential [18,19] and long-time production. Regulations worldwide are centred on PFAAs. As an example, Table S2.2 reports existing guidelines and directives for natural surface water, groundwater and drinking water in the European Union, Canada, Australia, and the U.S. Regulations are nevertheless shifting towards a grouping approach in order to speed up the dismissal of non-essential PFASs, and, ultimately, PFASs as a whole [20], but several challenges remain. Fluorine-free replacements (hereinafter “alternatives”) are either unavailable or not yet industrially feasible for commercial products because of the high costs [21–23], though sector-specific alternatives exist [24]. As a result of the phase-out process of long-chain PFASs, industry increased the production of short-chain PFASs and developed substitutes of the “legacy” PFASs commercialized for decades, that can be referred to as “novel” PFASs [25]. These substitutes are significantly less known and investigated, not regulated, and potentially harmful for human health. An overview of their most known representatives and their effects on human health is provided in section 1.1. Because of tighter PFASs regulation in Western Countries, PFAS production has shifted to Eastern countries [26], particularly China. Even though PFASs manufacture in China started in the 1980s [27], production of PFASs in China rapidly increased [28] and the country became the world’s most important producer of PFOS, PFOA and related chemicals [19,29–32]. Eastern China is the most important emission source for PFOA-related fluorochemicals [33,34]. The Country was reported to be responsible for 63% and >90% of the global production of PFBS and PBSF (perfluorobutanesulfonyl fluoride) in 2015 [35]. Despite the ratification of the Stockholm Convention and a grant from Global Environment Facility (GEF) to support the reduction of production and use of PFOS, its salts and perfluorooctanesulfonyl fluoride (PFOSF), the country has become the only known manufacturer of PFOS [27,32]. No mandatory concentration threshold for natural and drinking water has been established in China at national level [36,37], as for the U.S., though single states have been developing their own drinking water standards [38,39] and a National Primary Drinking Water Regulation for PFOA and PFOS has been proposed by U.S. EPA [40,41]. Available data suggest that the concentration of PFOS and PFOA in China’s population is increasing as opposed to the global population [42], but the comparison of PFASs occurrence in China with respect to Western Countries is limited. Different PFASs profiles and environmental concentrations could affect the efficacy of strategies, methods, materials, and technologies for the removal of this class of contaminants from the environment. Moreover, even the definition of PFASs is still under revision, potentially broadening the class and shaping new directions for management strategies [1,2].

The purpose of this work is to highlight hotspot and background concentrations in representative Western Countries (essentially European Countries, the U.S., and Canada) and in China, taken as long-time and recently industrialized countries, respectively, with special attention to novel and short-chain PFASs as new challenges in PFASs management. Hereafter, the terminology and classification used by Buck et al. and the OECD will be adopted [3,4,18] as suggested by expert scientists and regulators [43,44]. Accordingly, abbreviations concerning PFASs chain length will refer to the number of carbons in the alkylated chain rather than the number of carbons in the molecule (e.g. PFBA= C4 PFCA and PFBS= C4 PFSA). Though short-chain PFAAs are themselves legacy PFASs, the term “substitutes” is henceforth used to indicate both short-chain and novel PFASs. Further information about PFASs nomenclature and

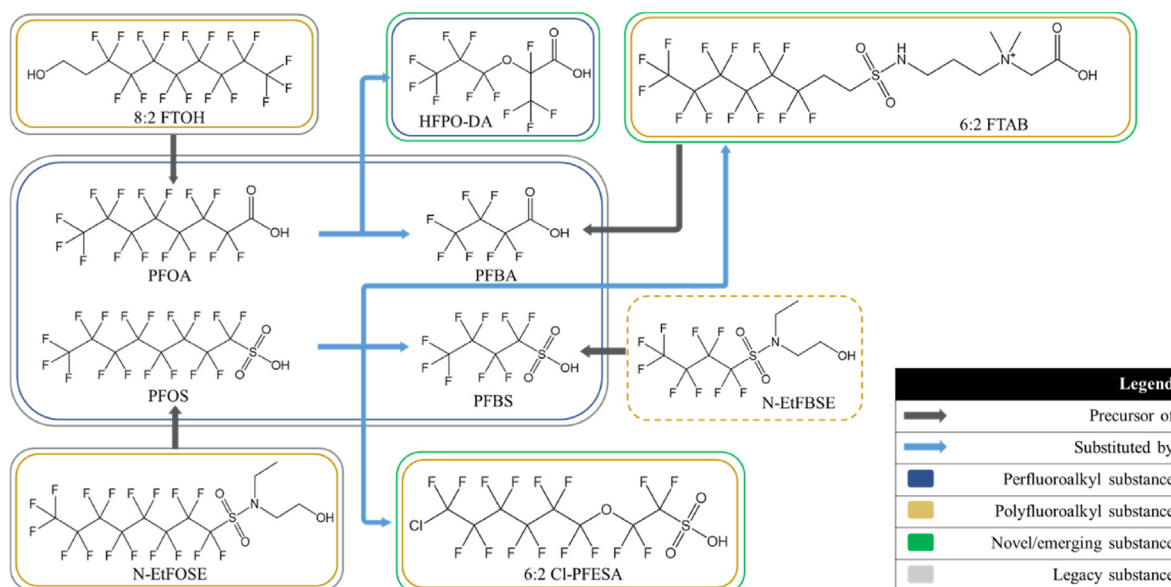
physical-chemical properties, as well as the acronyms used, can be found in the supplementary information (sections S1–S2). The following introductory subsections report relevant information on short-chain and novel PFASs (definition, health effects, knowledge gaps).

### 1.1. Short-chain and novel PFASs: overview and causes of concern

As previously mentioned, information on effects, physico-chemical properties, and the production of substitutes is scarce [45–48]. PFBS, PFBA, and PFHxA are among the most important short-chain PFAAs substitutes of PFOS and PFOA [49,50], while perfluoroalkyl ether carboxylic and sulfonic acids (PFECAs and PFESAs) are among the most relevant and studied new-generation PFASs [48,51]. Novel PFASs are characterized by structural modifications such as the addition of oxygen bridges or the substitution of fluorine atoms with other halogen or hydrogen atoms within the alkylated portion of the molecule [21,47]. For example, PFESAs and PFECAs are characterized by the presence of ether groups within the perfluorinated chain to ease the breakdown of the molecule into short-chain PFAAs, which are less bioaccumulative than the long-chain homologues [48,52]. Other than substitutes, several PFASs have been recently identified in environmental surveys. These substances are usually identified as “emerging” PFASs and encompass substances that are not necessarily newly introduced or short-chained. In fact, since the Stockholm Convention’s restrictions, the production of some already-existing PFASs has increased dramatically. For example, 6:2 Cl-PFESA (widely used as mist suppressant in metal plating processes) and OBS (sodium p-perfluorooxynonoxylbenzenesulfonate, an additive of foams used to enhance oil extraction) have been commercialized since the 70s–80s and are widely used in China [53,54], to the point that they can dominate the PFASs profile [55]. Likewise, 6:2 FTAB started being investigated recently although its use in aqueous film-forming foams (AFFF) has been known for a long time [56]. As such, the distinction between legacy and newly introduced PFASs is often unclear. The pathways that lead to the formation of perfluoroalkyl substances from polyfluoroalkyl precursors are related to both production (telomerization and electrochemical fluorination) [3] and direct or indirect (i.e., through the formation of intermediates) degradation in the environment [7]. Fig. 1 shows some key degradation precursors (in a wide range of matrices and conditions) and substitutes in specific application fields (introduced recently and a long time ago) of PFBA, PFOA, PFBS, and PFOS. Hereafter, “novel PFASs” is adopted in this work to indicate emerging PFASs as well as what has been reported so far in the literature as novel. Table S1.1 shows the chemical structure of selected novel PFAS.

Despite lower bioaccumulation potential in comparison with the long-chain counterparts, the higher hydrophilicity combined with the environmental persistence of short-chain PFAAs possibly results in higher mobility and in the permanent exposure of aquatic organisms with respect to the long-chain homologues (i.e., poorly reversible body burdens) [50]. An increase in relative concentration of short-chain PFAAs over time was highlighted in household products [61] as well as drinking water [62]. This is consistent with the short-chain PFASs not being as effective as long-chain ones in bestowing water and oil repellence so that larger quantities of short-chain PFASs need to be applied to achieve similar performances [25]. Furthermore, the lower affinity of short-chain PFAAs towards commercially available sorption materials represents a relevant problem for PFASs removal [63,64].

Estimation of physicochemical properties of novel PFASs by means of in-silico tools and in-vivo toxicity tests suggest similar environmental persistence with respect to the legacy counterparts



**Fig. 1.** Selected examples of 1) poly- and perfluorinated precursors and substitutes of PFAAs and 2) legacy and novel/emerging substances [3,57–59]. N-EtFBSE (N-ethyl perfluorobutanesulfonamidoethanol) is reported in a dashed line being a possible precursor of PFBS [60].

[46,65]. A chlorinated PFESA (8:2 Cl-PFESA) was calculated to have the longest half-life in humans among PFASs [66]. Novel PFASs like 6:2 FTS, 6:2 Cl-PFESA, HFPO-DA (GenX), HFPO-TA, HFPO-TeA, have been linked to comparable or higher genotoxic and endocrine-disrupting effects in humans than their homologues (PFOS and PFOA) [67,68]. Negative effects on human semen quality and adverse birth outcomes were observed for F-53B constituents (Cl-PFESAs) [69–71], other than alteration of lipid metabolism and endocrine disorders [71]. 6:2 FTS, 6:2 Cl-PFESA, and PFOPA were found at  $\mu\text{g/L}$  level in maternal and cord serum samples of women living near an e-waste dismantling site in China [72]. Other organisms showed toxic effects after exposure to cC6O4 [73], Nafion byproduct 2 [74], OBS [65], and GenX [75], pointing to a toxic potency similar to that of long chain PFAAs [65,75].

Analysis of total, adsorbable and extractable organic fluorine (TOF, AOF and EOF) as well as quantification of targeted PFAAs before and after chemical oxidation (i.e. TOP Assay) of polyfluoroalkylated precursors of PFAAs outlined that targeted PFASs analysis alone can lead to an underestimation of PFASs occurrence [76–81] as novel and unknown PFASs can represent a considerable fraction of total PFASs in environmental samples.

## 2. Occurrence of PFASs in wastewater, surface water and drinking water

Publications on PFASs occurrence in wastewater (WW), surface water (SW), groundwater (GW) and drinking water (DW) of Western Countries and China published between 2011 and 2022 have been reviewed and compared. The complete dataset can be found in Table S3.1 – S3.4. Section S4 details the number of studies found for each type of water for each region. Studies focused on ultra-short chain PFAAs, which are defined as PFAAs with  $C \leq 2$  (PFCAs) and  $C \leq 3$  (PFSAs) perfluorocarbon atoms, were purposely excluded from this review, since despite their increasing environmental relevance [82–85], there is still little data on their occurrence with the exception of trifluoroacetic acid (TFA), the sources of which (both natural and xenobiotic) are too numerous to allow a reliable comparison with other PFASs [84,86]. As each publication

examined a different number of analytes, the range of total PFASs is specified as  $\sum_{\text{min-max}} \text{PFASs}$  (where “min” and “max” indicate the lowest and highest number of analytes, respectively). Boxplots and histograms shown in this section are based on the average concentrations reported in the literature and only show results from target analysis (Figs. 2–5, Tables 2–5), while relevant aspects of untargeted and/or suspect-screening analysis are presented in section 3. Data used for the reported boxplots is shown in SI (part 2). If not present and where possible, the average and median concentration of single and total PFASs were calculated by assuming both absence of detection (“n.d.”) and concentrations below the limit of detection and quantification (<LOD and <LOQ) equal to zero. If concentration values were only reported as min-max range, midrange values were calculated and used instead of the average values. Outliers were not excluded to better represent the high variability of the reported concentrations. Other than total PFASs, PFBA, PFBS, PFOS and PFOA were selected as individual compounds to be compared as they are extensively analysed in the literature and represent the most used long- and short-chain PFAAs. In the case of short-chain PFCA, average PFHxA concentrations were included (if available) in the boxplot whenever those of PFBA were not reported, and PFPeA was also included in the boxplot related to SW hotspots of Western Countries (Fig. 4F) because of the fewer reported concentrations of PFBA and PFHxA. Where possible, to better compare PFASs presence in hotspots vs less contaminated areas (as regards SW, GW, DW) and in municipal vs industrial WWTPs (WW), concentrations of PFASs in samples with different levels of contamination were separately grouped to generate the boxplots. For example, concentrations related to 6 municipal and 2 industrial WWTPs examined by Houtz and collaborators were separately accounted for [87], similarly to SW samples analysed in urban, rural, and hotspot areas by D’Agostino and Mabury [88]. Urbanized, rural and remote areas vs highly urbanized/industrialized areas vs hotspots (e.g., fluorochemical production areas, AFFF-impacted sites) were reported and compared in the case of SW. In the case of DW, PFASs in bottled vs. tap water were also reported and evaluated.

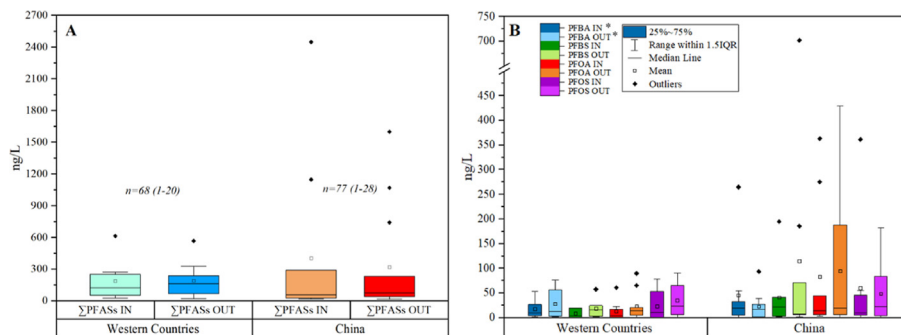


Fig. 2. Average concentrations of total PFASs (A) and selected PFAAs (B) in the influent WW (IN) and effluent WW (OUT) of municipal and mixed WWTPs in China and Western Countries. “n” = number (total and range) of examined WWTPs; \* = or PFHxA.

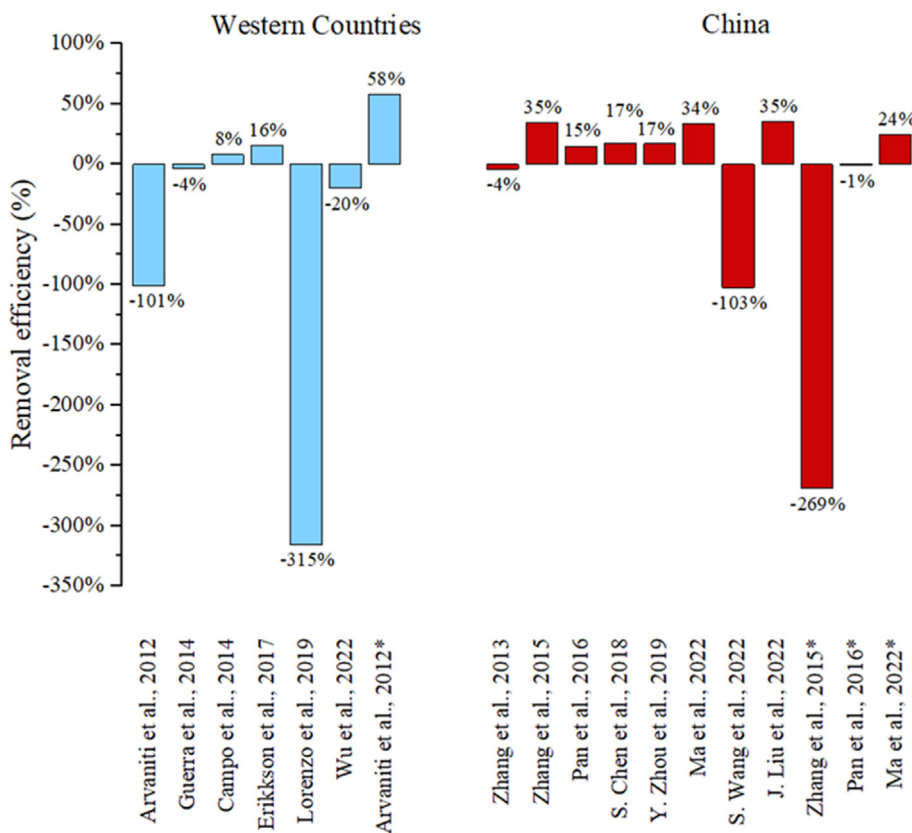


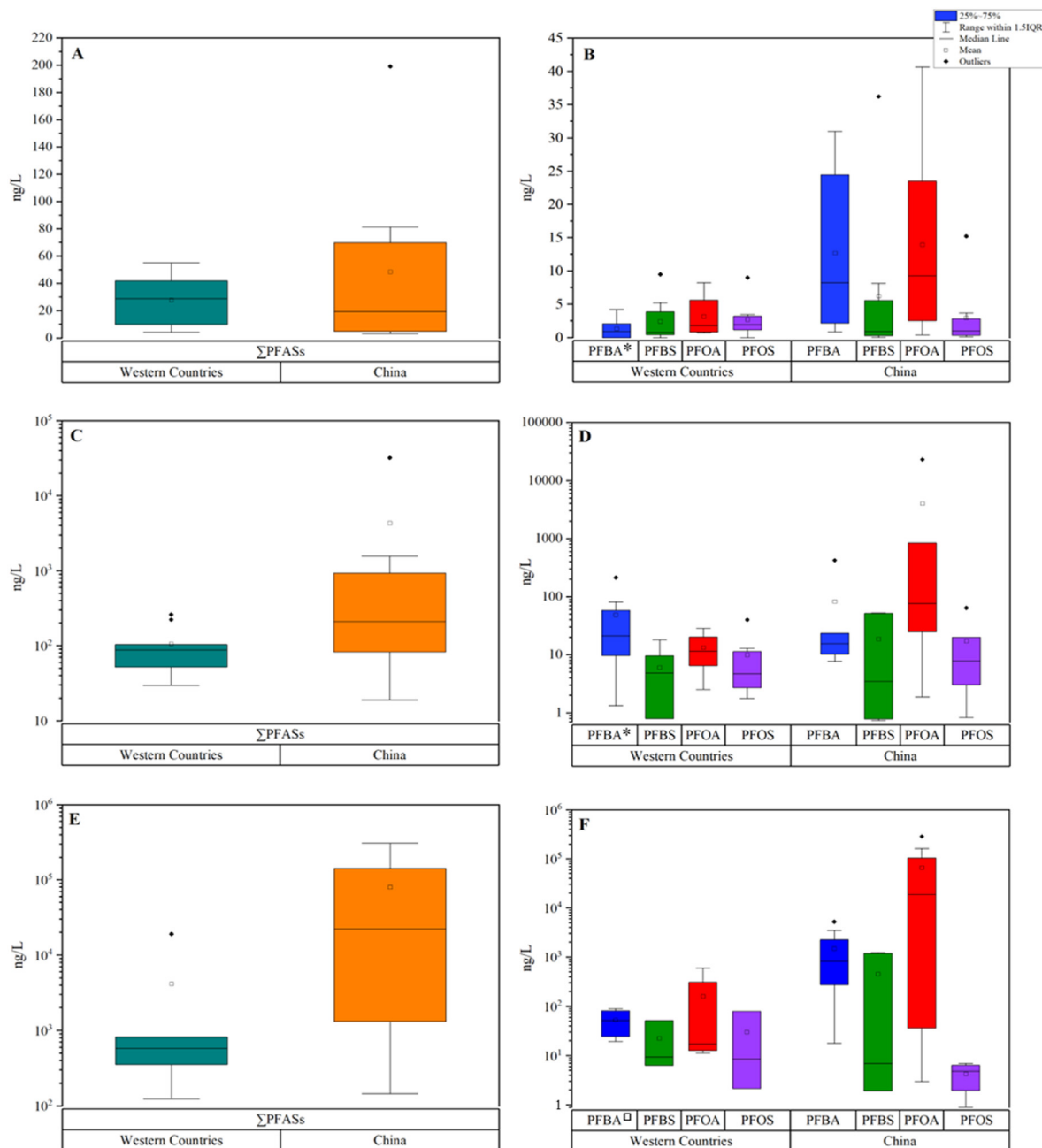
Fig. 3. Removal efficiency of total PFASs calculated as  $((C_{in} - C_{out})/C_{in}) * 100$ , where  $C_{in}$  = concentration in the influent WW,  $C_{out}$  = concentration in the effluent WW. \* = industrial WWTPs.

### 2.1. PFASs in wastewater

Wastewater treatment plants (WWTPs) represent a major source of PFASs, especially when serving industrial plants, factories, or industrial hubs, where PFASs are used in the manufacturing process or produced on-site [89]. The number of WWTPs considered in the 29 selected papers is considerably variable (2–20 in Western Countries, and 2–28 in China), as is the extension of the geographical areas considered, number of analytes, sampling frequency, and method. Concerning Western Countries, five publications involved the examination of industrial treatment plants [90–93] and a fluorotelomer factory [80], and the other two included WWTPs that receive non-municipal WW (from an airport, a military base, and a firefighting training area) [87,94]. Some

WWTPs receive mixed municipal and industrial WW [95,96]. Where not specified, the plants were considered to be receiving mixed WW [97]. As far as China is concerned, 10 publications reported on municipal WW, 4 on mixed WW, 3 on WWTPs serving industrial areas and 2 on WW of fluorochemical production plants.

As far as Western Countries are concerned, C9–C12 PFCAs were usually investigated and C9–C12 PFCAs were usually involved in the investigations, in addition to a few precursors, especially perfluoroalkyl sulfonamides (e.g., FOSA) and related molecules (e.g., FOSAA, EtFOSAA), fluorotelomer compounds (FTs), and polyfluorinated phosphate esters (PAPs), other than perfluorophosphonic and perfluorophosphinic acids (PFOPAs and PFPiAs). As shown in Fig. 2A, concentration of  $\sum_{4-41}$  PFASs detected in inlets and outlets of municipal and mixed municipal-industrial

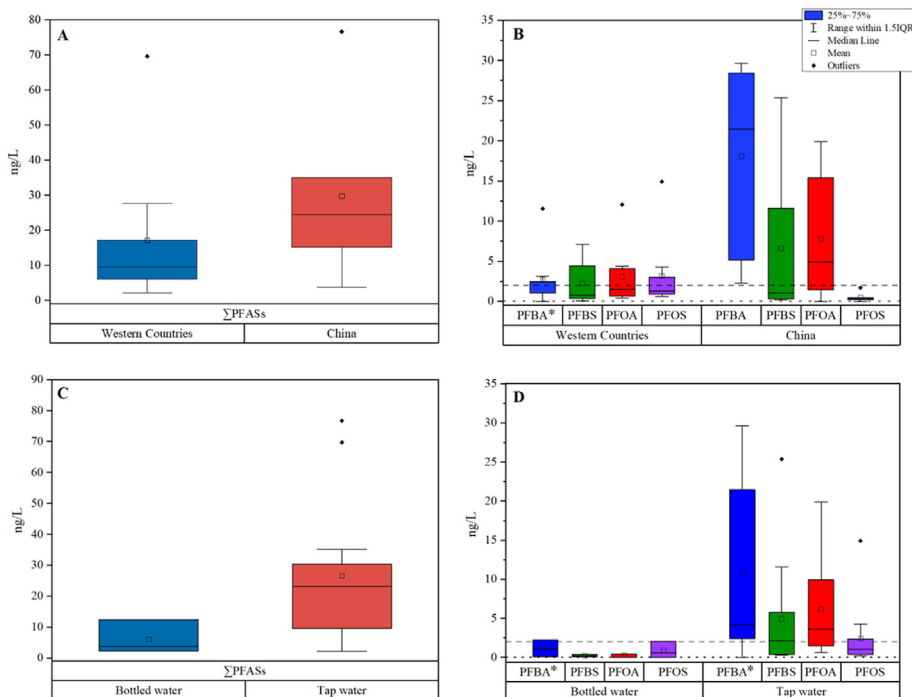


**Fig. 4.** Average concentration levels of total PFASs and selected PFAAs in SW of remote, rural, and urban areas (A, B), industrial areas (C, D) and hotspots (E, F). Histograms from C to F are reported in a log<sub>10</sub> scale; \* = or PFHxA; □ = or PFPeA.

WWTPs ranged from tens to hundreds of ng/L [87,91,95–98]. In industrial plants ( $\sum_{4-15}$ PFASs, Table 1) average concentrations between  $1 \times 10^3$ – $3 \times 10^3$  ng/L are typically involved [87,90,91,93].  $\sum$ PFASs in fluorochemical plants and firefighting training areas can reach  $1 \times 10^5$ – $1 \times 10^6$  ng/L concentrations both in China and Western Countries [28,80,94,99]. Some plants that treat waters from industrial hotspots also showed concentrations in the mg/L range [80,91,94], as in China [100], though mass flows can differ significantly and may even be negligible [91]. Relative contribution of C > 10 PFAAs in municipal WWTPs appeared to be more relevant in earlier surveys [95,101], but decreased significantly with time, as opposed to C4 and C6 PFAAs [87,90,96,98], with few exceptions [97].

The mean and median concentrations of total PFASs and

selected C4–C8 PFAAs in influents and effluents of WWTPs of mainly municipal origin (Fig. 2, A–B) are similar, with the main exception of PFOA. A reasonable explanation of this discrepancy might rely on the ongoing production of PFOA and related substances in China as opposed to Western Countries, where it has been phased out: a recent review brought out significantly higher concentrations of PFOA in DW in areas hosting polytetrafluoroethylene (PTFE) manufacturing plants [37]. Similarly, the mean value of PFBS is higher in China despite the comparable median value in both China and Western Countries. This is likely due to the contribution of particularly high concentrations in mixed WWTPs [104,105] which may reflect the high PFASs content in industry-related wastewater since PFBS is extensively produced and used in China [35]. PFBS was the predominant PFAS in the vicinity of



**Fig. 5.** Average background concentrations of total (A) and selected (B) PFASs in DW of Western Countries and China, and overall average concentrations of total (C) and selected (D) PFASs in tap and bottled water. The dashed and dotted lines (C,D) represent the EPA health advisory concentrations for PFBS and PFOS-PFOA, respectively. \* = or PFHxA.

**Table 1**

Average concentration of total PFASs ( $\Sigma$ PFAS), PFBA (or PFHxA), PFBS, PFOA and PFOS in WWTPs linked to industries (I), fluorochemical production plants (F) and downstream a firefighting training area (FFTA). “In” = influent WW; “out” = effluent WW; \* = midrange value.

		$\Sigma$ PFAS		PFBA/PFHxA		PFBS		PFOA		PFOS		Ref.
		IN	OUT	IN	OUT	IN	OUT	IN	OUT	IN	OUT	
ng/L												
Western Countries	I		667*		112.5*				432*			[90]
	I		1645*									[87]
	I	4890.5*	499.5*	103*	41*	4273*	258*	51*	21.5*	4*		[93]
	I	165.5*	719*									[92]
	I	64.27		9.41		1.95		8.26		7.07		[102]
	F	10600006*	5142080*	543	8625			580	40350			[80]
China	FFTA		7740539		4914		8384		6474		24535	[94]
	I	2100	4200	23	27	3.8	6	16	25	1300	2700	[103]
	I	590	520	21	18	0.36	0.24	52	47	8.2	2.2	[103]
	I	20410500	738000									[100]
	F	80945	30436	2849	6271	2	3	532	3630	n.d.	1	[28]
	F		546211		129		1		19232		6	[99]

Chinese fluorochemical industries [106–108] and increasing levels of PFBS over time were reported [106,107,109]. Therefore, though the highest (midrange) concentration of PFBS in industrial WWTPs is related to Western Countries [93], and PFOS was the dominant PFAA in relation to specific industrial productions [103] (as shown in Table 1), the shifting trend of long-towards short-chain PFAAs is evident even for PFBS.

The removal efficiency of total PFASs (Fig. 3) and of C4–C8 PFAAs (Table 2) is also overall comparable between Western countries and China, although highly variable. PFASs are not efficiently removed by WWTPs (approx. 8–60% of removal), and their concentration can substantially increase from the influent to the effluent (up to approx. 260–300%). PFASs behaviour can be considerably affected not only by the treatment itself but also by solution chemistry parameters like pH that can influence the electrostatic interaction between PFASs and organic matter, and, therefore, the separation

from the aqueous phase. It is also known that polyfluorinated compounds can be degraded down to PFAAs during the treatment process [7,110], and contribution of precursors can vary as well [92].

In China, C4–C12 PFCAs and C4, C6 and C8 PFASs were systematically investigated, and only a few precursors (long-chain sulfonamides, phosphate diesters, FTOHs, chlorinated PFESAs) were examined via target analysis (8 papers). Similarly to Western Countries, concentration of  $\Sigma$ PFASs in municipal and mixed industrial/municipal WWTPs (Fig. 2) revolves around tens to hundreds of ng/L [28,104,111–117], up to a few thousands of ng/L [100,105,118]. Generally, C4 and C8 PFAAs dominate the PFASs profile, like in Western Countries, but the concentration and composition profile of PFASs in municipal WWTPs are quite variable [112,115,118]. Although the transition towards short-chain PFASs was confirmed by several authors [89,119,120], long-chain compounds can still represent the most abundant PFASs [115].

**Table 2**  
Removal efficiency of selected PFAAs calculated as  $((C_{in}-C_{out})/C_{in}) \times 100$ , where  $C_{in}$  = concentration in the influent WW,  $C_{out}$  = concentration in the effluent WW. \* = industrial WWTPs.

Location	PFBA (or PFHxA)	PFBS	PFOA	PFOS	Ref.	
China	-10%	0%	-23%	0%	[111]	
	65%	-69%	-18%	-54%	[105]	
	17%	-1%	-18%	37%	[104]	
	15%	23%	1%	46%	[112]	
	-117%	-382%	-31%	13%	[113]	
	27%	-19%	32%	50%	[100]	
	15%	-1829%	-460%	-20%	[105]*	
	-15%	5%	98%	-99%	[104]*	
	-10%		12%		[114]*	
	Western countries	-8%	-203%	33%	-17%	[101]
		-507%	0%	-18%	53%	[96]
			-48%			[86]
		-185%	-28%	7%		[95]
		2%	-28%	-54%	-44%	[98]
	8%		-1821%	-213%	[97]	
	-125%	-71%		98%	[95]*	

**Table 3**  
Selected novel PFASs in surface waters of Western Countries and China that occurred at concentrations >1 ng/L. MDL = method detection limit; MQL = method quantitation limit; LOD = limit of detection; LOQ = limit of quantitation; n.d. = not detected; □ = background concentration; \* = concentration detected in oilfields.

PFAS	Western Countries			China			Ref.
	Range	Average	Median	Range	Average	Median	
HFPO-DA	0.59–8.75	1.73		<LOQ- 143.7	3.18		[136]
	0.49–107.6	40.31		n.d.- 3825	357.82		[137]
	<10-4560	<10-631	<10-304				[162]
	0.03–3600	270.53		<MDL- 3800	358.26		[138]
					1.09		[163]
HFPO-TA	<LOQ- 4.33	0.35		0.29–5.7			[164]
				<LOD-47.1	3.54		[134]
				1.61–2060			[125]
				<LOQ-34.8	1.07		[136]
				0.46–3.4			[164]
6:2 Cl-PFESA				3.69–68500	2.29–1923	2.20–1960	[125]
				0.01–52.2	3.39		[136]
					3.5		[163]
				0.28–3.2			[164]
				0.02–8.37			[119]
6:2 FTS	n.d.-2.86 3.9				34.1		[134]
				1.52–15.6	4.55–7.40	2.55–5.95	[125]
							[137]
				<LOD- 15	0.81		[152]
				0.91–7.6			[149]
OBS				<MQL-3200	6.9 <sup>□</sup>	5.5 <sup>□</sup>	[164]
					305*	144*	[65]
cC6O4	<40-290			1.7–28			[164]
							[161]

**Table 4**  
Selected novel PFASs in groundwater of Western Countries and China that occurred at concentrations >1 ng/L; n.d. = not detected. RL = reporting level.

	Western Countries			China			Ref.
	Range	Average	Median	Range	Average	Median	
HFPO-DA				<0.06–8.93	3.6	2.58	[178]
6:2 Cl-PFESA				0.02–18000			[171]
				0.04–13.41	3.43	0.4	[178]
				0.17–1.83			[168]
6:2 FTS	150*						[152]
				0.32–8.54			[168]
				0.04–14.3	2		[169]
	<RL-17		4.7				[180]
	2800–175000						[172]
8:2 FTS	0.15–5900		700				[133]
				<0.06–117.5	1.36	0.87	[178]
				n.d.-0.58	0.12		[175]
					1.1		[169]
		18000–27000					[172]

**Table 5**  
Selected novel PFASs in drinking water of Western Countries and China that occurred at concentrations >1 ng/L. MDL = method detection limit.

PFAS	Western Countries		China		World		Ref.
	Range	Average	Range	Average	Range	Average	
	ng/L						
HFPO-DA	<10-4560	631					[160]
	1.4–8	4.4					[183]
4:2 FTS	<MDL-4.63						[11]
6:2 FTSA					6.3	0.3	[187]
5:3 FTCA					39	1.2	[187]

Occurrence in industrial WWTPs hovers around a few hundred to a few thousand ng/L, as for Western Countries, with the exception of a WWTP serving an electroplating industry in China that showed concentrations similar to those found in fluorochemical manufacturing areas ( $1 \times 10^5$ - $1 \times 10^6$  ng/L) [100].

Overall, concentrations of total and single (PFBA, PFBS, PFOA and PFOS) PFAAs in Western Countries and China are comparable. A 10-

year temporal trend analysis in Australian WW brought out the progressive replacement of long-chain PFCAs and PFASs with short-chain PFCAs, in addition to polyfluorinated PFOS substitutes [121]. Though the shifting process towards short-chain homologues in China [122] seems to be slower or less uniform [89] due to e.g. areas with different economic development, attention towards PFASs occurrence in Chinese water treatment facilities is increasing [112].

Despite the phase-out process and the decreasing use of C8 compounds [90], C8 PFAAs still contribute remarkably to the total PFASs concentration both in municipal and industrial WWTPs. A study on wastewater from different industrial sectors in Korea highlighted the prevalence of long-chain homologues in the metal industry (PFDoDA and PFOA were the dominant homologues in 2012 and 2018, respectively) [89]. Some sectors still rely on the use of long-chain PFASs (especially C8 PFAAs), whose relative contribution in industrial plants was higher than short-chain homologues both in Western Countries [80,90,91] and in China [122,123]. Samples collected at different stages of electroplating process showed different relative compositions between production workshops using electrolytic degreasing (dominance of PFHxS, PFHxA and PFBA) vs chemical degreasing (dominance of PFOA, PFHxA and PFOS) [123]. PFOA and PFOS were the most relevant PFASs in terms of relative contribution, concentration, and detection frequency in the river surface waters of Tianjin, where >200 electroplating factories and >50 aircraft manufacturing enterprises were present [124]. PFOA represented the dominant PFASs in samples of river water immediately downstream of one of the major Chinese fluoropolymer manufacturing facility, followed by HFPO oligomers (HFPO-DA and HFPO-TA) [122,125]. The ongoing release of PFOA in China at fluoropolymer production sites can be attributed to the ongoing use of ECF-derived ammonium penta-decafluorooctanoate (APFO) in the manufacturing process [122]. Therefore, PFOS and PFOA use in industrial processes in combination with the long estimated environmental half-lives [126] can explain the substantial levels of C8 PFAAs that are still detectable in WWTPs and in the environment.

Recently introduced PFASs such as PFECAs and PFESAs, as well as fluorotelomer and sulfonamide-based PFAA precursors were found at concentrations comparable to C4–C8 PFAAs in municipal and industrial WWTPs [86,93,100,103,118]. For example, two out of ten industrially-impacted WWTPs in Michigan showed significant levels of 6:2 FTS both in influent (~31 and ~250 ng/L) and effluent (~43 and ~1200 ng/L) [92]. The production of substitutes and precursors in fluorochemical manufacturing areas can overcome that of legacy compounds [3,80,125]. However, the occurrence and fate of PFAAs precursors and of recently introduced poly- and perfluoroalkyl substitutes in WWTPs is understudied. Out of 29 papers, 16 only examine PFAAs, 9 papers up to 3 additional compounds (mostly FOSAs and related compounds like EtFOSA, EtFOSAA), and only 3 studies examined the occurrence of e.g. GenX, ADONA, or OBS in wastewaters. Hence, the role of WWTPs as secondary source of precursors and substitutes in the environment should be further investigated. There are numerous variables and operating conditions that drive the environmental fate of PFASs in WWTPs, like the type of treatment, the residence time in the aeration tanks (i.e. the contact time with microorganisms capable of degrading precursors or the physical separation from the aqueous phase by means of air fractionation), presence of organic matter and ions (which influence the adsorption on the solid phase as well as on adsorption materials), etc. PFAAs are known to be extremely resistant to degradation, which is presumably negligible under the operating conditions of full-scale plants. PFECAs with no -O-CF<sub>2</sub>- moiety were not degraded during TOP assay [127], while e.g. ADONA was degraded to PFMOPra, suggesting a different amenability to chemical and, possibly, microbial attack, but more studies are

needed to elucidate their susceptibility to biodegradation. 6:2 and 8:2 Cl-PFESAs were barely affected by both aerobic biodegradation tests in soil and by heat-activated persulfate oxidation [128]. Moreover, little is known about the hydrophobicity and partitioning behaviour in sludges and biosolids.  $K_d/K_{oc}$  values need to be determined and studied under different conditions. It is noteworthy that the partitioning behaviour of PFASs towards sludges and biosolids can significantly change with respect to variables like treatment methods, and medium composition parameters among which level of proteins > organic matter > lipids [129]. PFAAs were also formed from zwitterionic and cationic precursors during chlorination and ozonation [130]. Reported studies show that substances like fluorotelomers can occur at comparable or higher concentrations than PFAAs [96], especially in manufacturing plants and hotspots [80,94]. FTOHs, FTSS, diPAPs decreasing concentrations from the influent to the effluent [28,80,96,114]. Although partitioning towards solid matrices and -with regard to FTOHs- volatilization must be considered, the biodegradation of such compounds (towards other polyfluorinated intermediates and PFAAs) has been proved in different matrices and conditions [7,59].

Three studies carried out in WWTPs serving industrial plants highlighted the significant contribution of polyfluoroalkyl substances and tentatively identified PFASs. The most extensive investigation of PFASs in WWTPs by means of targeted analysis [80] reported the fate of 51 PFASs throughout a WWTP serving a fluorochemical facility, showing an overwhelming dominance of the fluorotelomer precursors over PFAAs both at the influent (approx. 2000–12640 vs  $5700 \times 10^3$ – $2200 \times 10^4$  ng/L, respectively) and effluent (approx.  $71 \times 10^3$ – $280 \times 10^3$  vs  $2350 \times 10^3$ – $7700 \times 10^3$  ng/L, respectively), even in terms of mass flow (0.6–8.6 g/day vs 647–2892 g/day). Additionally, both oxidative conversion of precursors and AOF analysis underlined how PFASs occurrence was still heavily underestimated. Similar observations were reported by Houtz et al. (2016) for both municipal and non-municipal WWTPs, again indicating a prevalence of short-chained PFAAs-precursors, and by Jacob et al. (2021) for electronic fabrication facilities where the total amount of PFASs in the diluted discharge samples were estimated to increase by a factor of 100 when including semi-quantified PFASs from nontarget analysis (i.e. from about 370 to 620 ng/L to 1400–78000 ng/L).

## 2.2. PFASs in surface water

Fig. 4 shows average concentrations of total PFASs and of C4–C8 PFAAs detected in areas with low anthropogenic pressure (i.e., averagely-poorly urbanized, rural, and remote areas) as well as industrialized/highly urbanized areas and hotspots (where PFASs are produced or have been used extensively and dispersed in the environment). As regards concentration, frequency of detection and relative abundance, C4–C8 PFCAs and C4–C8 PFASs represent the dominant PFASs. On average, levels of total PFASs and selected PFAAs are higher in China, especially near industrialized areas and hotspots, in line with the role of China as production leader of PFASs. Among PFAAs, PFOS shows remarkably higher concentrations in hotspots of Western Countries than those of China. Though it is reported that China is the only manufacturer of PFOS and related compounds, the production of this chemical has possibly been reduced with time in compliance with the Stockholm Convention and the above-mentioned GEF [131]. Moreover, not only PFOS has been massively produced in Europe and the U.S. in the last decades, but some authors highlighted that many impacted sites like military bases [62,132] and areas linked to extensive use of AFFFs [92,133] in Western Countries still show relevant concentration of legacy compounds like PFOS, that was used in AFFF formulations. PFOA, on the other hand, is significantly more

widespread in China (as highlighted for WW) where it is the dominant PFAS together with PFBA in hotspots, in line with the production of PFOA and its related chemicals. Some authors pointed out the widespread presence of PFOA with respect to shorter PFCAs homologues in Chinese rivers impacted by fluorochemical factories [134], different from Dutch and German rivers [135]. A worldwide investigation on 24 selected PFASs in surface water of China, the U.S., the U.K., Sweden, Germany, the Netherlands, and Korea showed how concentration ranges in China were comparable to those detected in industrialized areas (approx. 20–300 ng/L), whereas the concentrations found in the other countries were similar to background levels of urbanized areas (approx. 16–75 ng/L) [136]. Similarly, lower concentration ranges of PFASs were detected in European rivers in comparison with Chinese rivers [137,138].

Highest concentrations of  $\sum_{12-38}$ PFASs in hotspots were found in China (roughly  $400 \times 10^3$ – $1860 \times 10^3$  ng/L) [125,137–139]. Concentrations ( $\sum_{12-21}$ PFASs) up to a few  $\mu\text{g/L}$  were found in rivers and river basins in highly urbanized and industrialized areas of Spain, Italy, Germany [140–143] and China [116,131,137], in a Finnish firefighting training area [133] and close to an oil industry in Tianjin [124]. Samples collected at a distance from the fluorochemical plants or hotspots show 1–2 orders of magnitude lower concentrations. In fact, maximum concentrations in industrialized areas can be often ascribed to a few samples taken nearby fluorochemical plants (or in river basins hosting such plants) [116,134,142,144], or to industrial water discharges potentially related, for example, to tannery and textile sectors [140,142]. Lower concentrations levels were detected in China around Tianjin (Bohai Sea) ( $\sum_{19}$ PFASs = 103–443 ng/L) [145], in rural areas of Eastern China ( $\sum_{17}$ PFASs = 7–489 ng/L) [144], and Taihu Lake ( $\sum_{21}$ PFASs = 96.3–330 ng/L) [146]. Slightly lower and comparable concentrations were found in Beijing surface waters ( $\sum_{17}$ PFASs = 2.9–222 ng/L) [147], Rhône river ( $\sum_{4}$ PFASs = 13–200 ng/L) [148], Ebro river basin (51.9 ng/L as average of  $\sum_{12}$  detected PFASs) [141], and urban areas of New York-Rhode Island ( $\sum_{14}$ PFASs = approx. 0.6–225 ng/L) [149] and Finland ( $\sum_{23}$ PFASs = 15–75 ng/L) [150]. Moderately or poorly urbanized areas show PFASs concentrations that are an order of magnitude lower with respect to urbanized/industrialized areas, like in Swedish rivers [151] and French Guiana [152] ( $\sum_{26}$ PFASs = 1–60 ng/L, and  $\sum_{20}$ PFASs = <LOD–77 ng/L respectively). In a survey of the Danube River across several Western Europe Countries, the highest PFOA concentration was 37 ng/L [153]. As far as remote areas are concerned, Tianchi lake (Tianshan mountains, China) was found to be contaminated with low levels of PFASs ( $\sum_{17}$ PFASs = 0.79–15.41 ng/L) [154], similarly to Finnish rivers ( $\sum_{23}$ PFASs = 0.53–42 ng/L) [150].

Several authors pointed out the increasing contribution of shorter chain and/or novel PFASs to the relative abundance with respect to previous studies [119,136,146,155–157] and/or the declining levels of long-chain PFASs [149]. This trend was also confirmed by PFASs in Alpine ice cores [158]. A survey of 19 PFASs carried out in a river impacted by the presence of fluorochemical manufacturing facilities (Fuxin, China) showed how PFBA and PFBS were the most frequently detected analytes and showed the highest overall concentrations [159]. Heydebreck and co-workers (2015) found lower PFBS concentration in the Lower Rhine River (Germany) with respect to a study from 2008 but ascribed such results to reduced discharge rates in wastewater. In general, there is evidence that the phase-out process of legacy long-chain PFASs is rapidly progressing [119,131,159], but long-chain homologues can still dominate the sample composition [131]. Estimates of PFASs mass-flows in rivers impacted by the presence of PFASs-producing and PFASs-using industries indicate a considerably higher

production in China [138].

Novel compounds and PFAAs precursors were scarcely investigated (e.g. only 12 publications monitored  $\geq 5$  novel PFASs and/or precursors). However, PFECAs, PFESAs, and C6 fluorotelomer sulphonates like 6:2 FTS were found in concentrations comparable to C4–C8 PFAAs both in China and Western Countries (Table 3). For example, GenX was found in American (<10–4560 ng/L), European (0.59–8.75; 0.03–3600; 0.49–107.6), Chinese (<LOQ–143.7; <MDL–3800; 0.29–5.7; n.d.–3825; 1.61–2060 ng/L) surface waters, as well as 6:2 FTS. PFECAs are increasingly used as substitutes of PFOA in synthesis and production of fluorochemicals [125,160]. OBS and X:2 Cl-PFESAs are only produced, used, and monitored in China, where they were frequently detected at relevant concentrations. For example, 6:2 Cl-PFESA was detected in all surface water samples in a highly industrialized area of Tianjin and showed concentrations comparable to those of PFOS and PFOA (0.16–22 ng/L), and the detection rate of 6:2 FTAB, 6:2 FTSA, and FOSA was approx. ~80%. 6:2 FTAB, FOSA, PFOSB, and PFOSAm reached concentrations of 1300, 110, 38, and 22 ng/L respectively nearby an oil industry [124]. The PFOA substitute cC6O4 was found in natural freshwater (<40–290 ng/L) during environmental monitoring of a contaminated area in northern Italy [161]. Hotspots in China are mainly related to industry and fluoropolymer/-telomer manufacture [131], where numerous substitutes occur at significant levels. Less economically developed areas show different PFASs patterns in comparison with the most industrialized parts [119].

### 2.3. PFASs in groundwater

GW contamination is of particular concern as it can be used as a source of drinking water. PFASs-polluted areas like AFFF-impacted sites, fluorochemical manufacturing areas and agricultural land amended with contaminated biosolids can act as a slow-releasing secondary source of PFASs towards the unsaturated and saturated zone for years.

Twenty-four papers were selected to discuss groundwater occurrence of PFASs (Table S3.3), including a publication from 2010 [165] and a technical report by a regional environmental agency in Italy [166]. Approx. half of the total publications are focused on China and the others on Western Countries.

With respect to non-industrialized or rural areas, the concentration range of total PFASs ( $\sum_{17-34}$  PFASs in China,  $\sum_{6-14}$  PFASs in Western Countries) is variable (from <LOD to a few tens to a few thousands ng/L) [144,152,156,162,165,167–169]. Nevertheless, highest concentrations can be ascribed to samples collected nearby hotspots [144,162,168].  $\sum_{10-12}$ PFASs in hotspots occurred in the range of thousands or tens of thousands ng/L [107,139,143,166,170]. PFASs levels in rural and non-industrial areas were 1–2 order of magnitude lower with respect to those found around a nearby fluorochemical production area and in an industrial site, respectively [144,168]. Highest concentrations were found in the U.S. (from 71 ng/L to ~4.4 mg/L as combined concentration of PFOS and PFOA), China ( $\sum_{10-43}$  PFASs up to  $\sim 3.7 \times 10^3$ ,  $15.6 \times 10^3$ ,  $26.7 \times 10^3$ , and  $270 \times 10^3$  ng/L), and Italy ( $\sum_{13-14}$  PFASs up to  $\sim 1.5 \times 10^3$ – $2.1 \times 10^3$  ng/L). High levels of PFOA (<0.02–9800 ng/L) and GenX (0.02–18000 ng/L) were also determined in the Netherlands [171].

The occurrence of the above examined PFAAs suggest a higher relative contribution of PFOS (with respect to the other PFAAs) in Western Countries, especially in hotspots, where highest concentrations were detected in AFFF-impacted sites [133,143,172]. PFOS and related compounds have been extensively produced and used in Western Countries prior to the worldwide ban and restrictions. Furthermore, PFOS has been reported to irreversibly bind to soil as opposed to PFOA and PFBS [173], which could partially explain the lower relative concentrations and detection frequencies of PFOS

with respect to other PFAAs in comparison with surface waters reported in some studies [159]. An appreciable difference in relative composition of PFAAs can be noted when confronting samples collected at different distances (<1 km, 1–4 km, 4–7 km, and 7–10 km) from a fluorochemical manufacturing facility, since long chain PFCAs and PFSA strongly decrease with distance as opposed to short-chain homologues [139].

C4 to C8 PFCAs and PFASs represent the most abundant and most frequently detected analytes [156,166,168,170,174]. A 24-fold PFBS increase (from  $\sim 0.9 \times 10^3$  to  $21.2 \times 10^3$  ng/L) in comparison with a 2009 survey in the Liaoning Province was reported, and PFBA was the dominant PFAS in many of the investigated sites together with PFOA [107]. C < 6 PFAAs showed the highest concentrations, relative abundance, and detection frequency (followed by PFOS and PFOA) in groundwater from non-industrial areas of the Jiangsu Province [168]. PFBS and PFHxS were the most frequently detected compounds in French overseas territories [152] and in the Loess Plateau of China [169]. Likewise, a 100% detection frequency of PFBA, PFBS, PFHxA, PFHxS and PFOA and a 96% and 92% detection frequency of PFPeA and PFOS around a perfluorosulfonates manufactory in Hubei province (China) was reported. PFBA and PFBS showed the highest detected concentrations [170]. Similar detection frequencies were reported in Tianjin (China), where C4 PFAAs were detected in all samples and dominated the compositional profile [156,163].

Other works reported different results, with long-chain PFAAs showing a higher relative abundance [169,175], which could be explained by local industrial use and/or production, as well as by the higher affinity of long-chain PFASs to the solid matrices that can lead to a slower leaching into the subsurface aquifers. Soil/aquifer geochemical composition and PFASs properties (i.e. PFASs behaviour, including adsorption onto soil particles) together with sampling depth can affect PFASs concentrations [176]. Furthermore, composition and concentration of PFASs in surface waters can vary seasonally, suggesting the role of precursors biodegradation and/or more intensive runoffs during wet seasons, i.e. a greater contribution of shorter, more hydrophilic PFASs to groundwater [145,177].

10 peer-reviewed papers and 2 reports investigated the occurrence of precursors and novel PFASs, essentially long-chain sulfonamides and related compounds (FOSAs, FOSEs), X:2 FTSS, X:2 Cl-PFESAs, HFPO-DA/TA, PFOPA, other than X:2 diPAPs, X:2 FTAs, X:2 FTUAs, phosphonic acids and perfluorophosphinates. Table 4 reports on selected novel PFASs found at substantial concentrations. Only two publications explored a wide suite of novel PFASs [169,175]. The investigation of 4:2,6:2, and 8:2 FTS, 6:2 and 8:2 Cl-PFESA, HFPO-DA, HFPO-TA, PFECHS, ADONA brought out detection frequencies of 86.7–100% for 7 of these novel compounds. 6:2 FTS showed the highest maximum concentrations among all analytes in French overseas territories (150 ng/L) [152] and occurred at comparable levels to PFCAs in the Jiangsu province in China (0.32–8.54 ng/L), where it showed a 100% detection frequency together with F-53B (0.17–1.83 ng/L) [168]. In the Loess plateau of China 6:2 FTS, Cl-PFESAs, ADONA, and HFPO homologues, were detected in 96.7–100% of the samples [169]. In the U.S., 6:2 FTS was found at much lower concentration than PFAAs (<0.5–1.2 and n.d.–4.3 ng/L, respectively) according to a recent review [162] but reached  $175 \times 10^3$  ng/L in groundwater beneath military bases and airports of Florida [172]. X:2 FTSS (X = 4,6,8) and 3:3 FTA showed detection frequencies between 23 and 52% and 85%, respectively [175]. In a survey carried out in Jianxi province 6:2 Cl-PFESA, 6:2 FTS and HFPO-DA were detected in 100%, 71.6% and 35.2% of samples, respectively. The same study estimated daily intakes relative to PFBS, 6:2 FTS and 6:2 Cl-PFESA similar to that of PFOS [178]. Estimated concentrations of butyl pentadecafluorooctanoate, among other substances, were far higher than those of any legacy

compound in the Danube River basin (106 ng/L) [179].

#### 2.4. PFASs in drinking water

PFASs are not completely removed in DW treatment plants and are consistently found in DW worldwide. It has been estimated that millions of people are exposed to PFASs via DW [37,155]. Background occurrence in terms of average concentration of  $\sum_{13-17}$  PFASs in tap and bottled water revolves around a few units to a few tens of ng/L [62,181–186] for both Western Countries and China (Fig. 5A). Comparable results in terms of median and average concentrations were found in >500 DW samples covering 66 Chinese cities (approx. 15 and 38 ng/L, respectively) [37]. Overall, PFBA, PFBS, PFOA, and PFOS represent the most abundant and/or the most frequently detected PFAAs, in accordance with the results of a study on the worldwide distribution of 29 PFASs in tap and bottled water showing how PFBA and PFBS contributed the most in terms of relative abundance [187]. Average and median background concentrations of C4–C8 PFCA and PFSA in Western Countries and China are similar (1–5 ng/L), except for PFBA in China (approx. 20 ng/L) (Fig. 5B). In this regard, a 4-years monitoring (2011–2014) of a developing fluorochemical industry in China showed a clear increasing trend in the occurrence (and, therefore, production) of PFBA with respect to PFBS and PFOA, which collectively represent the dominant PFASs throughout the monitoring period [131]. Short-chain PFAAs (especially PFBA, PFBS, PFHxA) contribution to  $\sum$ PFASs increased with respect to previous surveys in Western Countries as well [62,160].

The lesser efficacy of typically applied treatment technologies (i.e. adsorption-based) towards short-chain compounds in potabilization processes could also represent a pivotal factor for the dominance of C4 PFAAs, which are highly hydrophilic [62,188,189]. For example, a relevant variability in the PFAS profile between different brands attributable to different water treatment processes was highlighted [174,186]. Tap water showed higher PFASs concentrations with respect to bottled water (Fig. 5, C–D), as brought out by an international investigation on the occurrence of 29 PFASs in drinking water [187]. Tap water samples collected nearby fluorochemical production sites mostly show concentrations up to a few tens ng/L [131,170,183], but reached a few  $\mu$ g/L in China and in the U.S. [37,182], such as source (raw) drinking water samples collected nearby the same kind of hotspots in other Western Countries [11,143,160,188]. A greater contribution of C8 PFAA (especially PFOS) was found with respect to C4 homologues in relation to the activities of firefighting training grounds and, presumably, widespread presence of military bases in the U.S. territory [62]. PFASs in the range of 0.1–502.9 ng/L (as averages of  $\sum_{18}$ PFASs) were detected in an extensive survey in China, with PFOA, PFOS and PFBA as the most abundant compounds. In fact, occurrence of short-chain PFASs and novel PFASs like F-53B and HFPO-TA increased considerably in recent years [37].

Only a few studies investigated the occurrence of novel PFASs: GenX was found in concentrations similar to PFOS within 25 Km of a fluoropolymer manufacturing facility in the Netherlands (1.4–8.0 ng/L) and immediately downstream of a fluorochemical production plant in North Carolina, with concentrations far higher than any other monitored PFASs (<LOQ–4560 ng/L) [160,183] but was not detected in bottled water across the United States [186]. 6:2 Cl-PFESA showed concentrations comparable to PFOS in a DWTP in Tianjin [163]. Only 4 publications investigated precursors (fluorotelomers and sulfonamides) [11,174,186,187]: 6:2 FTSA showed the highest maximum concentrations among all analytes in tap and bottled water (3.6 and 1.3 ng/L, respectively) [187], while 5:3 FTCA, an intermediate in the biodegradation of 6:2 FTS which is widely used in firefighting foams [96], showed among the highest average

concentrations [187]. The assessment of PFASs presence in drinking water is of utmost importance as it represents a particularly important direct exposure pathway to the biota. Concentrations thresholds and guidance values in drinking water regulation on PFASs has substantially changed over the years. According to the previous U.S. EPA health advisories for PFOA and PFOS, the previous indications in the EU Drinking Water Directive (DWD) for PFOS, PFOA and other PFASs, as well as existing limits in Canada and Australia (Table S2.2), PFASs presence in the reviewed literature is of little concern for public health and only PFNA reference levels exceeded [11,160,172,182,190] the Canadian screening values (20 ng/L) [191]. However, the most recent DW health advisories established by U.S. EPA [40] for PFOS and PFOA (20 pg/L and 4 pg/L) would systematically be exceeded both in tap and bottled water. A similar situation could take place even for PFBS (health advisory concentration: 2 ng/L) (Fig. 5 B-D, where the dashed and dotted lines represent the EPA health advisory concentrations for PFBS and PFOS-PFOA, respectively). The revised EU DWD [192] has fixed a limit of 100 ng/L in DW as sum of C4–C13 PFCAs and PFSAs, which is more easily achievable under the current emission scenarios. In addition, a threshold of 500 ng/L (“Total”) for the sum of all other PFASs was also included, but the methods to be used for the monitoring of this parameter and possible target analytes has not yet been defined. As inorganic and organic fluorine deriving from other widespread fluorochemical pollutants (e.g. fluoropharmaceuticals, pesticides) could lead to a significant overestimation of fluorine from PFASs, a TOP Assay-based approach is preferable: this assay (where PFSAs and PFCAs precursors are oxidized to PFCAs by hydroxyl radicals) provides a quantitative estimate of a fraction of PFAAs precursors and is relatively easy to carry out [169]. The method protocol of TOP Assay has been improved to encompass a broader spectrum of final degradation products for the target analysis (like ultrashort-chain PFCAs) [193] and additional PFAAs precursors have been assessed for their oxidative yields to PFCAs [194]. However, the method still presents non-negligible drawbacks, namely reproducibility and difficulty to ensure the total oxidation of all precursors (i.e. underestimation of the total PFASs) [194].

It was highlighted that more data on remote areas of China are needed [37]. This is also true for Western Countries, for which more peer-reviewed literature on PFASs in DW should be produced as well.

### 3. Discussion and recommendations

As shown by reviewed data, PFASs concentrations in natural waters of China and Western Countries are overall comparable although higher in China where environmental emissions are expected to increase with time due to the geographical shift of PFASs production from Western to Far Eastern Countries. In fact, as regards SW hotspots (especially those affected by PFASs production sites) and DW, the reported results suggest that Chinese population might be locally exposed to higher concentrations than population of Western Countries [37,42].

The comparable concentrations in the influents with respect to the final effluents of WWTPs, as well as the occurrence of PFASs in DW, indicate the worldwide inadequacy of potabilization and wastewater treatment technologies. WW, SW, GW and DW exhibit background concentrations up to some hundreds of ng/L, while hotspot concentrations are largely variable and range from hundreds ng/L to some mg/L. C4, C6, and C8 PFAAs are the most frequently detected compounds with the highest concentrations.

Novel and short-chain PFASs have become globally ubiquitous pollutants as much as the legacy long-chain PFASs [48]. Short-chain PFAAs occur at similar levels than C8 homologues, which are still

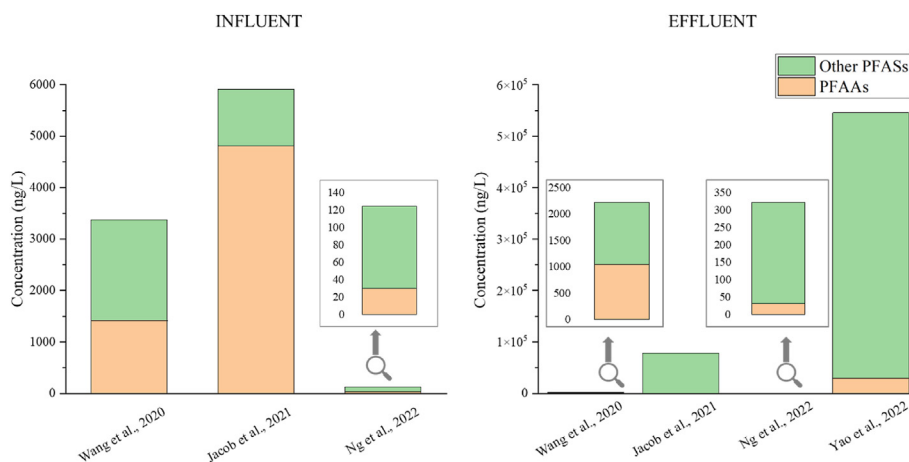
widely spread in the environment due to residual ongoing production, leakage from hotspots, and environmental persistence. However, PFASs other than C4–C12 PFAAs are still poorly investigated despite their ubiquitous presence in water, even at significant distance from fluorochemical production areas [136,137,168]. A broader suite of analytes (including at least C4 and C8 PFAAs precursors and selected new-generation PFAAs that showed widespread presence like GenX and 6:2 FTS) should be systematically included in environmental surveys.

Over the last 5–6 years, investigations carried out to determine non-regulated PFASs by both targeted and untargeted mass spectrometry analysis increased significantly [80,93,94,96,118,119,195] together with analysis of organic fluorine and TOP Assay [80]. The histograms reported in Fig. 6 show the average or median concentration of the frequently monitored PFAAs vs the sum of PFASs other than PFAAs [179] or vs that of PFASs detected via untargeted and suspect-screening methods. The latter allowed to identify 48–133 substances among the considered publications, of which 21–90 at level 3 or above according to the confidence-based classification method proposed by Schymanski and coworkers [196], and 6–40 PFASs classes; 8–37 PFASs substances and 6 homologous classes were reported for the first time. In many cases PFASs identified via explorative methods significantly contributed to the total PFASs concentration, even outweighing PFAAs. Of particular interest is a comparative examination of novel PFASs found in German and Chinese rivers hosting industries and fluorochemical factories [135]. The number of PFASs detected in the effluent of fluorochemical production sites increased by 1300% (Alz River) and 130% (Xiaoqing River Basin) with respect to upstream samples. The comparison of carbonyl-based and PFECAs homologue classes in the same rivers highlighted the dominance of short-chain substances in the Alz River, and of longer-chain substances in the Xiaoqing River Basin (except for PFdiCAs). Noticeably, untargeted and suspect/screening analysis was not applied in the reviewed papers on DW.

More systematic application of untargeted and suspect screening analysis would significantly improve the exposure assessment of humans and ecosystems to PFASs. This is confirmed by the evidence that the contribution of PFASs other than the known ones has increased in human blood serum over time [78].

As structure and physical-chemical properties of many PFASs likely to be found in the environment are not known, the “essential use” regulatory approach [20] is fully justified, and it is imperative to minimize the exposure to PFASs which requires the optimization of the identification and quantification strategy [8]. To achieve this goal, the following aspects should be taken in full consideration.

- i) More substances should systematically be included in target analysis, starting from novel compounds already found in the environment (e.g., PFECAs, PFESAs, unsaturated fluorotelomer sulfonates). Even though a high number of substances can potentially fall under the definition of PFASs, including e.g. pharmaceuticals and pesticides, the compounds of interest are possibly much less numerous. Useful computing tools based on consistent databases exist to support the analysis and identification of suspect compounds. For example, the PubChem Classification Browser [197] collects several databases that can be filtered out according to e.g. PFASs uses, structures, patents, or PFASs actually commercialized/produced. Moreover, with respect to the 2018 PFASs definition by OECD, experts from the major global producers of fluorochemicals proposed a restricted list of 256 (out of 4730) “commercially relevant” (i.e. currently produced compounds and related production impurities, ingredients, and degradation products) PFASs [198]. This



**Fig. 6.** Concentrations of PFAA and estimated concentrations of PFASs detected by explorative methods in the influent (left) and effluent (right) of WWTPs in China [99,118], the US [93], and Europe [179].

underlines, *inter alia*, the needed support by actual producers and industrial stakeholders for the management of PFASs.

- ii) When performing environmental surveys, it is advisable to pay full attention to the characteristics of the surrounding area, *i.e.* presence of nearby industrial activities, and to use existing peer reviewed literature and databases to identify PFASs of potential interest that could be used in specific industrial sectors [5,199]. The integration of untargeted analytical techniques, fluorine measurements, and assays based on the oxidation of precursors should be instrumental given the role of WWTPs as a source of PFASs (as such and as transformation products of precursors). Special attention should be given to WWTPs serving fluorochemical production facilities and PFASs-using activities, for which site-specific, targeted and more frequent monitoring activities should be pivotal to understand sector-specific shifting trends towards newly introduced compounds. For frequent PFASs surveys, TOP assay coupled with inorganic, organic and total fluorine analysis should at least be included to gain more realistic knowledge of PFASs occurrence, with special regard to DW and source DW that was analysed only via targeted analysis in the reviewed articles. Moreover, DW and GW should be studied more extensively, especially in Western Countries.
- iii) The occurrence of precursors, substitutes, as well as other poorly monitored compounds brings out the need to cope with a wide range of physical-chemical properties, and to set criteria to prioritize substances. The use of *in-silico* modelling tools has proven to be effective to help categorize and prioritize (*i.e.* individuate representative compounds) inventoried substances according to predicted properties that are relevant for risk assessment (*e.g.*  $K_{ow}$ , bioconcentration factor, vapor pressure). Nevertheless, it was highlighted that more experimental data is needed to train and validate such models [16].
- iv) Some studies underlined the relation between the number and/or type of PFASs detected (especially novel PFASs) and other variables like land use, co-occurrence of other recurring man-made chemicals [180], as well as gross domestic product (GDP) [200]. Such variables can provide useful information on PFASs exposure: for example GDP has been used as an indicator of the consumption of PFAS-containing products, that are so far more widely used in economically

developed areas. A stronger correlation between per capita daily emissions and environmental concentration of emerging PFASs was found with respect to legacy PFASs [118], and a positive correlation was found between concentration of total PFASs and GDP [111,118]. However, in urbanized areas with higher GDP where more efficient treatments of DW and WW are applied, lower levels of total PFASs were found [184]. McMahon and collaborators ranked 57 potential predictor variables for PFASs occurrence in groundwater. Co-occurrence of volatile organic carbons and pharmaceuticals were among the most important ones and give valuable insights both to chemical mixtures to which humans are potentially exposed to and to possible common sources of these chemicals. For example, as reported by the authors, concomitant presence of PFASs and pharmaceuticals is associated with, among others, WW effluents and landfill leachate [180].

#### 4. Conclusions

The results presented in this work show that the concentration of PFASs in Western Countries and China is quite comparable in industrial, urban, and remote areas. Nevertheless, concentrations of PFAS in China are overall higher, especially in hotspots. Therefore, it is reasonable to assume that Chinese population is locally exposed to higher concentrations than the population of Western Countries. Background concentrations in WW, SW, GW and DW can reach some hundreds of ng/L while concentrations in hotspots can reach a few mg/L. C4, C6 and C8 PFAAs are reported to be the most important compounds in terms of relative concentration and frequency of detection. An effective assessment and management of background and hotspot-related PFASs contamination must consider both old-generation PFASs, which still occur widely, and their substitutes *i.e.* short-chain and novel PFASs. The latter represent a relevant and increasing fraction of the total PFASs recorded in the examined environmental matrices. To this regard, short-chain and novel PFASs are still insufficiently investigated, and total PFAS present in the aqueous environment is significantly underestimated. The limited number of regulated substances introduces a systematic bias in the evaluation of the exposure to and of the occurrence of PFASs. As thousands of substances fall in the PFASs classification, it is necessary to introduce criteria to prioritize substances, develop robust *in-silico* predictive models, include

more novel PFASs as target analytes, and apply TOP assay-like based techniques to have a more realistic view of environmental occurrence of and human exposure to PFASs.

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### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.emcon.2023.100228>.

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