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Research Article

Occurrence, fate, and remediation for per-and polyfluoroalkyl substances (PFAS) in sewage sludge: A comprehensive review

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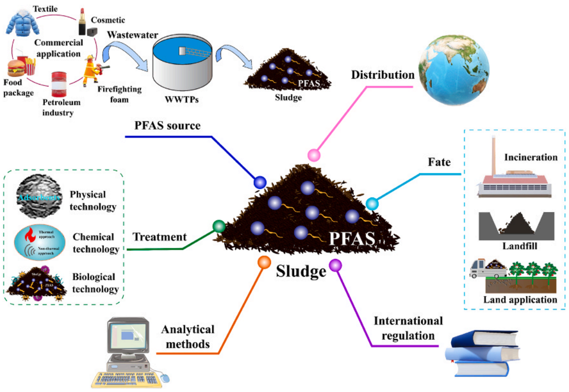
HIGHLIGHTS

- PFAS migration pathways from sources to the environment in sludge are depicted.
- Regulations for PFAS in sludge are summarized globally.
- Analytical methods for PFAS in sludge are discussed.
- Biological techniques are promising for PFAS remediation in sludge.

GRAPHICAL ABSTRACT



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ABSTRACT

Addressing per-and polyfluoroalkyl substances (PFAS) contamination is an urgent environmental concern. While most research has focused on PFAS contamination in water matrices, comparatively little attention has been given to sludge, a significant by-product of wastewater treatment. This critical review presents the latest information on emission sources, global distribution, international regulations, analytical methods, and remediation technologies for PFAS in sludge and biosolids from wastewater treatment plants. PFAS concentrations in sludge matrices are typically in hundreds of ng/g dry weight (dw) in developed countries but are rarely reported in developing and least-developed countries due to the limited analytical capability. In comparison to water samples, efficient extraction and cleaning procedures are crucial for PFAS detection in sludge samples. While regulations on PFAS have mainly focused on soil due to biosolids reuse, only two countries have set limits on PFAS in sludge or biosolids with a maximum of 100 ng/g dw for major PFAS. Biological technologies using microbes and enzymes present in sludge are considered as having high potential for PFAS remediation, as they are eco-friendly, low-cost, and promising. By contrast, physical/chemical methods are either energy-intensive or

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linked to further challenges with PFAS contamination and disposal. The findings of this review deepen our comprehension of PFAS in sludge and have guided future research recommendations.

Nomenclature

PFAS	Per-and polyfluoroalkyl substances.
WWTPs	Wastewater treatment plants.
PFCA	Perfluorocarboxylic acid.
PFSA	Perfluorosulfonic acid.
PFOA	Perfluorooctanoic acid.
PFOS	Perfluorooctane sulfonate.
PFNA	Perfluorononanoic acid.
PFDoA	Perfluorododecanoic acid.
PFDA	Perfluorodecanoic acid.
PFHpA	Perfluoropentanoic acid.
PFUnA	perfluoroundecanoic acid.
PFHxA	Perfluorohexanoic acid.
PFPeA	Perfluoropentanoic acid.
PFBA	Perfluorobutanoic acid.
PFBS	Perfluorobutane sulfonate.
PFPeS	Perfluoropentanesulfonic acid.
PFHxS	Perfluorohexane sulfonate.
n:2 FTOH	n:2 fluorotelomer alcohols.
N-EtFOSA	N-ethyl perfluorooctane sulfonamide.
N-EtFOSE	N-ethyl perfluorooctane sulfonamidoethanol.
USA	United States of America.
PFOSA	Perfluorooctane sulfonamide.

Abbreviations	Meaning.
SPE	Solid phase extraction.
WAX	Weak anion exchange.
HLB	Hydrophilic-lipophilic-balanced.
LC/(-)ESI-MS/MS	Liquid chromatography-electrospray ionization tandem mass spectrometric.
GC	Gas chromatography.
6:2 FTS	6:2 fluorotelomer sulfonate.
7:3 FTCA	7:3 fluorotelomer carboxylate.
8:2 FTS	8:2 fluorotelomer sulfonate.
n:2 diPAP	n:2 fluorotelomer phosphate diester.
F^-	Fluoride ions.
PFMeUPA	4-(Trifluoromethyl)hexafluoropent-2-enoic acid.
MeU-C4c	2-(Trifluoromethyl)acrylic acid.
MeU-C5d	4,4,4-Trifluoro-3-(trifluoromethyl)crotonic acid.
6:2 FTUCA	2 H-perfluoro-2-octenoic acid.
2 H-PFHpA	2 H-perfluoropentanoic acid.
2 H-PFHxA	2 H-perfluorohexanoic acid.
5:2 FTOH	5:2 fluorotelomer secondary alcohol.
n:2 FTCA	n:2 fluorotelomer carboxylate.
6:2 FTUCA	2 H-perfluoro-2-octenoic acid.
5:3 FTCA	5:3 fluorotelomer carboxylic acid.
8:2 FTUA	2 H-perfluoro-2-decenoic acid.

1. Introduction

Per-and polyfluoroalkyl substances (PFAS) are a group of fluorinated chemicals that contain a minimum of one fully fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it) [70]. The carbon-fluorine (C-F) bond in the molecular structures of PFAS provides increased thermal stability and degradation resistance, making them highly useful for industrial and commercial applications. By 2018, the global market had identified nearly 4700 PFAS variations [69], and the United States of America (USA) alone utilizes over 600 chemicals commercially [88]. Both point and nonpoint sources release PFAS into the environment [24]. Stationary and discrete facilities such as industrial factories, wastewater treatment plants (WWTPs), firefighting training sites, and landfills, are among the point sources. In contrast, nonpoint sources include the use of industrial and consumer products and atmospheric transport [24]. PFAS, with their wide application, high persistence, and potential for bioaccumulation, have become widespread in various environmental matrices, including aqueous and solid matrices [83,91]. This has raised concerns about their impact on human health, causing such issues as neurodevelopmental disorders, weakened immune defensive systems, adverse pregnancy outcomes, and increased cancer risks [113,114,55].

Among the various pathways that PFAS undergo, WWTPs act as transit points from commercial use to natural environments [33]. Numerous studies have investigated the occurrence of PFAS and treatment methods in wastewater as well as subsequent water and soil systems within wastewater-receiving areas. PFAS in environments pose ecological and health threats, such as surface water and soil contamination, bioaccumulation in aquatic organisms and plants leading to ecological imbalances, and entry into human through the food chain [12,14,46]. Several reviews have discussed the fate and treatment approaches of PFAS in wastewater systems [55], and the detection of PFAS in aquatic systems [83]; and summarized the physicochemical properties of PFAS and their remediation technologies in soil [64]. However, limited attention has been given to PFAS in sludge generated by WWTPs.

PFAS with greater hydrophobicity are absorbed into sludge during wastewater treatment processes [100]. Occurrence of PFAS in sludge has been documented globally such as Australia [66], Germany [80], the USA [91], and China [58], with concentrations varying from tens to thousands of ng/g dry weight (dw) [66]. Collected sludge is considered biosolids after undergoing appropriate treatments such as digestion, lime stabilization, composting, and heat treatment, to meet regulatory requirements [4,62]. The reuse and disposal of biosolids introduce PFAS into the environment such as agricultural lands, which make them a crucial source of discharge into the environment [66]. Recent analytical advancements have enabled in-depth investigations of PFAS in sludge [71]. The fate of PFAS during sludge stabilization has become a research hotspot for ensuring the safety of biosolid reuse, with various treatment technologies and regulations being proposed [109,110]. Additionally, regulations have been implemented by several countries to restrain PFAS concentrations in commercial products, biosolids, and soil. However, there is a lack of detailed summaries regarding the occurrence, detection, international regulations, and fate of PFAS in sludge systems.

This review conducted a comprehensive assessment of PFAS in sludge matrices, with the following critical objectives: (1) presenting analytical methods for PFAS; (2) identifying PFAS occurrence, transformation, and fate in sludge; (3) describing the associated toxicological risks and international regulations; and (4) discussing remediation technologies in detail. This study provides an in-depth analysis of removal efficiencies and mechanisms during sludge stabilization. The review provides an overview of the latest information regarding PFAS in sludge and identifies areas for further research into PFAS remediation technologies.

2. Analytical methods for the detection of PFAS in sludge

2.1. Overview

Scarce studies have consolidated the quantification of PFAS in sludge matrices, although the detection of PFAS in aquatic systems has been widely documented and discussed [83]. This is because PFAS are closely integrated with extracellular polymeric substances in sludge, making accurate identification and quantification challenging [100]. For accurate analysis, several procedures are essential, including sample collection and preservation, sample preparation, and instrumental analysis (Fig. 1), details of which are summarized in the following section.

2.2. Collection and preservation

For PFAS analysis, appropriate sample collection and preservation are crucial in minimizing losses and preventing contamination. Sludge samples are collected using pre-cleaned equipment and then transferred into polyethylene or polypropylene tubes (or bottles) [90]. Glass and fluoropolymer containers are strictly prohibited as glass can easily adsorb PFAS and fluoropolymer containers can introduce background contamination, causing inaccurate analysis [55]. Containers collected during the entire process are typically pre-treated with methanol and purified water. Following collection, samples are homogenized and stored at − 20 ℃ in the freezer for up to 90 days until analysis [90]. PFAS are highly stable under appropriate preservation conditions with limited decay [5].

2.3. Sample preparation

Sample preparation aims to release, capture, concentrate, and purify embedded PFAS in sludge matrices before analysis. The general workflow for sample preparation includes pre-treatment, extraction, and clean-up (Fig. 1).

Before extraction, sludge samples are generally freeze-dried and finely ground to create a normalized dry weight basis for accurate results [66]. Sludge and biosolids cannot exceed a maximum weight of 0.5 g dw [90]. Additionally, it is crucial to spike the unscreened samples with the native standard solution to the mid-level calibration point concentration or 3–5 times the background concentration for screened samples [90]. Afterward, organic solvents such as basic methanol solution are used with the aid of ultrasonication or homogenization to release embedded PFAS into the solvents, followed by centrifugation to collect the supernatants [33]. This technique is currently the most popular method for PFAS extraction in sludge and biosolids.

Since extraction techniques are not selective for PFAS, a clean-up process is vital to eliminate interference from other matrix substances and enhance quantification performance. Currently, a range of adsorbents have been used independently or in conjunction for extract cleanup. These adsorbents include octadecyl-functionalized silica (C18), weak anion exchange (WAX), hydrophilic-lipophilic-balanced (HLB), graphitized non-porous carbon (ENVI-Carb), primary and secondary amine-functionalized silica (PSA), and bare silica [33,56,59,71]. For specific properties of the adsorbents, Ozelcaglayan et al. [71] conducted a comparison of the performance of four adsorbents (WAX, C18, PSA, and ENVI-Carb) in cleaning up biosolid extracts. Their findings indicate that utilizing a combination of different adsorbent types improved clean-up performance. Nonetheless, decreasing the adsorbent amount may result in increased recovery efficiency due to PFAS adsorption on the adsorbents. Recovery values of PFAS increased significantly from 40–100% to 80–180% when the amount of PSA and C18 decreased from 1000 mg to 200 mg [71]. Additionally, due to the significant variations in sludge and biosolids, it is necessary to modify the adsorbent formulation to achieve an optimal trade-off between chromatogram quality and PFAS recovery. Activated carbon particles are utilized to further remove potential matrix constituents from the extract eluent before analysis [58,90].

2.4. Instrument analysis

Mass spectrometry has become a popular and convenient method for identifying and quantifying PFAS [37]. Numerous studies have reported that ionic PFAS, particularly for PFSA and PFCA, can be separated through C18 compatible guard columns with mobile phase and detected using liquid chromatography-electrospray ionization tandem mass spectrometric (LC/(-)ESI-MS/MS) with a detection limit at the

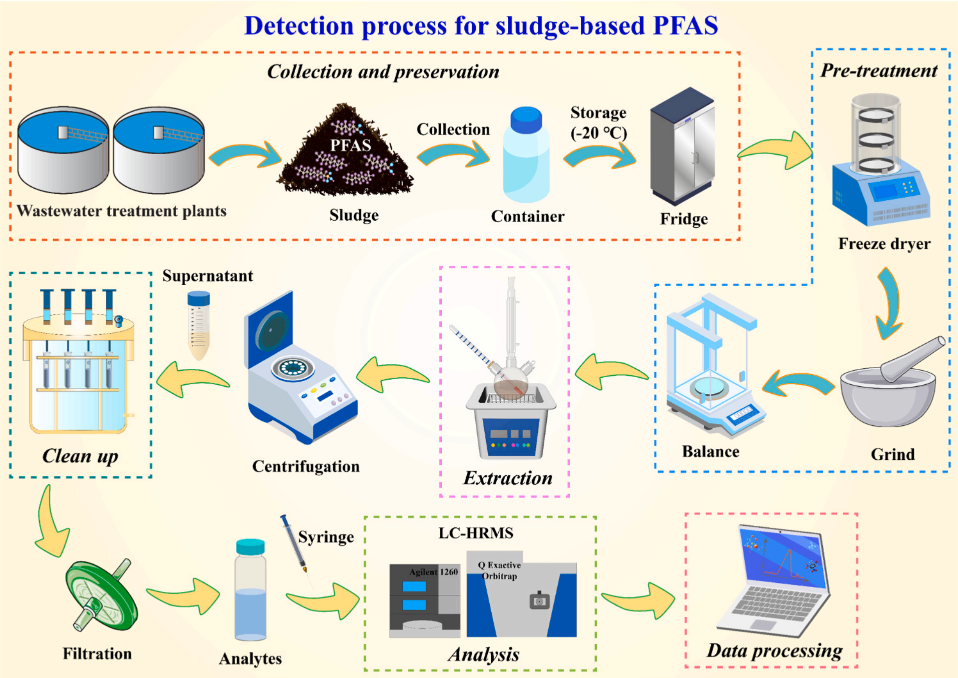

Fig. 1. Detection process of PFAS in sludge matrices.

nanogram level [71]. The injection volume of 2.0 μL is the default for LC-MS-MS [90]. Gas chromatography (GC)/MS is suitable for volatile, semi-volatile, and neutral PFAS [37]. Shen et al. [78] found that some ionic PFAS such as PFCA are also detected by GC/MS after derivation, with better separation ability and less instrument-related pollution. Despite the superior PFAS separation techniques of GC/MS, LC-MS-MS is more prevalent than GC/MS [36]. The longer and more complicated preparation process, limitations in detecting volatile and neutral PFAS, and unreproducible data through GC/MS create potential drawbacks, compared to LC-MS-MS [36].

Even though both LC-MS-MS and GC/MS are extensively used, they exhibit similar drawbacks, including time-consuming and costly analysis, and inability to enable in-situ detection [36]. Furthermore, the identification of certain PFAS precursors (e.g., N-EtFOSA) and PFAS transformation products cannot be achieved through conventional quantification techniques due to insufficient sensitivity and the unavailability of reference standards. Accurately quantifying PFAS in highly complicated matrices poses challenges due to the limited availability of isotopically-labeled PFAS analogues. Therefore, further research is recommended to develop new techniques for rapid and precise detection, including non-targeted screening strategies.

2.5. *Non-targeted screening*

Non-targeted screening strategies typically employ high-resolution mass spectrometry (HRMS) systems for PFAS analysis in various environmental matrices [63]. HRMS systems include ion mobility spectrometry, quadrupole time-of-flight mass spectrometry, and fourier-transform ion cyclotron resonance mass spectrometry, which can collect MS/MS spectra to quantify the structures and concentrations of PFAS sensitively and precisely [78]. For instance, ion mobility spectrometry separates ions in the gas phase by shape, size, and charge state. The use of ion mobility spectrometry coupled with LC/MS identified 23 new PFAS homologous series [63]. Che et al. [18] employed LC-HRMS to identify the structures of PFAS intermediates and revealed aerobic defluorination pathways of PFAS. Despite the structural proposal for unknown analytes using HRMS, further structural confirmation is required. When HRMS is combined with other analytical techniques such as inductively coupled plasma mass spectrometry (ICP-MS) and ^{19}F- nuclear magnetic resonance (^{19}F NMR) spectroscopy, non-targeted screening strategies also provide valuable tools for quantifying mixed fluorine substances and confirming isomers [78].

However, despite advances in non-targeted screening methods, challenges remain [78]. For instance, ^{19}F NMR has limited sensitivity, which restricts its use in trace-level analysis. ICP-MS faces difficulties in inefficient fluoride ions (F^-) formation and isobaric interferences. Furthermore, data processing in HRMS systems is complex and time-consuming. To overcome these difficulties and broaden the analytical range, various alternative techniques have been proposed, including fluorine K-edge XANES spectroscopy and combustion ion chromatography (CIC) [76]. These methods provide improved sensitivity and data analysis capabilities, contributing to a more comprehensive understanding of PFAS contamination and the advancement of effective mitigation strategies.

Overall, non-targeted screening strategies, coupled with advanced analytical techniques, have greatly enhanced the ability to detect, identify, and quantify PFAS in complex environmental samples. Further research and development on sensitivity, precision, and data processing abilities will facilitate the development of effective strategies to mitigate PFAS contamination.

3. Occurrence of PFAS in sludge matrices

3.1. *Distribution of PFAS in sludge matrices*

With advances in analytical methods, a variety of PFAS have been globally identified in sludge, including North America, Europe, and Asia [33,5,77]. PFAS in sludge are mostly divided into perfluorocarboxylic acids (PFCA, $C_nF_{(2n+1)}CO_2H$) with carboxyl groups (-COOH) and perfluorosulfonic acids (PFSA, $C_nF_{(2n+1)}SO_3H$) with sulfonate groups (-SO_3H) based on their functional groups (Table 1). Moreover, PFAS can be further classified by carbon chain length as short- ($C \leq 7$) and long-chain PFCA ($C > 7$), and short- ($C \leq 6$) and long-chain ($C > 6$) PFSA [70].

The concentrations and variety of PFAS in sludge vary among and between continents (Table 1). Of the limited data available, PFAS concentrations in sludge were the highest in Switzerland (up to 2854 ng/g dw), followed by Thailand (up to 2231 ng/g dw), and lowest in Nigeria with a maximum PFAS concentration of 2.57 ng/g dw. Perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) were found to be the primary compounds of PFAS in sludge from most countries (Table 1), representing the typical subgroups of PFCA and PFAS, respectively.

In North America, Venkatesana and Halden (2013) investigated PFAS concentrations in biosolids collected from 94 WWTPs in 32 states and the District of Columbia in 2001. Their findings indicated that PFOS (308–618 ng/g dw) was the predominant PFAS, followed by PFOA (11.8–70.3 ng/g dw) and perfluorodecanoic acid (PFDA) (6.9–59.1 ng/g dw). No significant difference ($P > 0.05$) in PFAS concentrations in biosolids collected in 2001, 2004, and 2007 was observed in the study [91], despite the 3 M company discontinuing the industrial production of PFOS and related compounds in the USA since 2000. This finding indicates that the effects of past production of PFAS may still be present in biosolids, demonstrating the persistent property of PFAS.

Armstrong et al. [5] observed stationary trends of 12 PFAS in biosolids collected from a municipal water resource recovery facility in the USA over an 8-year period between 2005 and 2012. The highest concentration of PFOA detected was up to 601 ng/g dw, followed by perfluorononanoic acid (PFNA) (<0.3–218 ng/g dw) and PFOS (<0.8–68.1 ng/g dw). The significant discrepancy in PFAS concentrations found in biosolids between the two U.S. studies probably because the broader sampling scope in the Venkatesana and Halden (2013), which may include industrial sources. Diverse treatment processes undergone by biosolids samples in the two studies may also contribute to the variations in PFAS concentrations. Although located in the same continent, limited data collected from 29 Canadian WWTPs demonstrated that PFOS was the most abundant among PFAS (<0.49–50.4 ng/g dw) in collected biosolids, followed by PFDA (0.11–53 ng/g dw) and PFOA (<0.07–23 ng/g dw) (Table 1). However, their levels are comparatively lower than those found in the USA. This discrepancy between the two countries could be attributed to differences in industrial practices, usage patterns, and regulatory measures.

In Australia and European countries, such as Germany, Denmark, Switzerland, and Greece, PFOS and PFOA were identified as the primary PFAS in sewage sludge with concentrations ranging from < 0.01 to 2440 ng/g dw and < 0.03 to 87 ng/g dw, respectively (Table 1). In Asian countries, such as China and Thailand, PFOS was discovered to be the major PFAS, with concentrations of 28–135 ng/g dw and 397–553 ng/g dw, respectively (Table 1). From the available data, PFAS concentrations in sludge samples from African countries like Nigeria and Kenya were relatively low, with PFCA and PFSA levels detected at less than 0.6 ng/g dw and 0.9 ng/g dw, respectively (Table 1). Notably, PFAS discharge and occurrence in sludge increase in highly urbanized and industrialized areas [55]. For countries with limited manufacturing output, the presence and abundance of PFAS are largely reliant on imported commodities [55], such as Australia, where 8% of the annual imported PFOS finally enter biosolids [66]. Data gap regarding PFAS concentrations in the sludge of least-developed and developing countries may be caused by relatively lower release concentrations/usage of PFAS and poor monitoring capabilities, considering the low-income and resource-poor conditions [79,86]. Inadequate access to high-technology equipment and insufficient funds for necessary chemicals are among the

Table 1
Concentrations of PFAS measured in sewage sludge and biosolids are given as a range (minimum-maximum), while mean concentrations are presented in parenthesis.

Continents	Countries	PFCA (ng/g dw)								PFSA (ng/g dw)		
		PFBA (C4)	PFPeA (C5)	PFHxA (C6)	PFHpA (C7)	PFOA (C8)	PFNA (C9)	PFDA (C10)	PFUnA (C11)	PFDoA (C12)	PFBS (C4)	PFHxS (C6)	PFOS (C8)
Europe	Greece[6]	-	< 0.26-45.2 (6.6)	< 0.22-61.5 (4.0)	< 0.30-16.4 (1.9)	< 0.36-19.4 (3.4)	< 0.38-13.5 (1.7)	< 0.26-9.6 (1.3)	< 0.05-5.5 (1.6)	< 0.46-9.8 (1.2)	< 0.06	< 0.06-18.3 (2.3)	0.58-16.7 (6.7)
	Denmark [13]	-	-	-	-	0.7-19.7 (0.4)	0.4-8.0 (1.5)	1.2-32.0 (7.2)	0.5-4.4 (1.2)	-	-	0.4-10.7 (3.6)	4.8-74.1 (18.4)
	Switzerland [2]	0.8-49	0.7-14	0.7-91	0.7-22	1.0-87	0.9-23	0.9-73	-	-	0.1-28	0.1-27	4.0-2440
	Spain (Navarro et al., 2011)	-	< 0.05-4.69 (1.46)	< 0.03-2.60 (0.42)	< 0.01-2.04 (0.28)	< 0.03-7.94 (2.85)	< 0.01-10.23 (1.23)	< 0.04-24.29 (5.41)	-	-	-	< 0.01-18.20 (1.46)	< 0.01-286.81 (63.99)
	Germany [80]	-	< 1-112 (3.8)	< 1-16.4 (1.0)	< 1-26.8 (0.5)	< 1-50.6 (6.5)	< 1-23.3 (1.8)	< 1-42.8 (4.1)	< 1-6.4 (0.7)	< 1-29.1 (1.7)	< 1-10.3 (0.4)	< 1-95.3 (1.6)	< 1-698 (23.1)
	Sweden [33]	< 0.6	< 0.1	0.3-8.2	< 0.3-1.6	< 0.3-44	N.D.– 39	N.D.– 69	N.D.– 120	N.D.– 22	< 0.1	0.1-2.5	0.6-26
North America	Canada [56]	< 0.48-3.0	< 0.28-6.0	0.17-4.65	< 0.08-1.53	< 0.07-11.5	0.09-4.72	0.11-23.4	0.19-7.49	0.19-6.09	< 0.14-3.48	< 0.06-2.43	0.49-50.4
	Canada [52]	< 2.06	< 2.06-14	< 2.06-8.3	< 2.06-5.2	< 2.06-23	< 2.06-20	< 2.06-53	< 2.06-7	< 2.06-10	< 4.11-11	< 4.11	< 4.11-27
	USA[5]	-	1.54-27.7 (8.3)	< 0.18-21.1 (7.7)	< 0.1-67.7 (5.1)	< 0.66-601 (23.5)	< 0.3-218 (25.1)	-	-	-	-	-	< 0.8-68.1 (22.5)
	USA[91]	1.2-3.2 (2)	1.8-6.7 (3.5)	2.5-11.7 (6.2)	1.2-5.4 (3.4)	11.8-70.3 (34)	3.2-21.1 (9.2)	6.9-59.1 (26.1)	2.8-38.7 (11.7)	4.5-26 (10.9)	2.5-4.8 (3.4)	5.3-6.6 (5.9)	308-618 (403)
Asia	China[58]	N.D.– 131	N.D.– 46.7	0.35-118	0.6-137	9.21-75.5	0.8-3.51	0.33-6.21	0.31-23.4	0.4-4.48	N.D.– 2.61	N.D.– 3.06	28.1-135
	South Korea[77]	-	-	1.16-4.47 (3.16)	0.68-1.35 (1.05)	5.24-10.89 (8.18)	3.86-12.88 (6.92)	10.39-13.14 (11.42)	67.91-111.66 (96.94)	3.75-6.51 (5.26)	0.98-36.97 (13.39)	0-1.66 (0.92)	4.55-61.67 (26.39)
	Thailand [49]	-	2.9-3.3	0.3-99.9	1.6-52.6	11.3-136.0	5.1-512.8	3.8-327.7	45.2-78.2	N.D.– 310.6	-	36.6-157.7	396.9-552.6
Oceania	Australia [66]	N.D.– 3.8 (0.8)	< 0.3-9.6 (2.0)	< 0.3-17 (2.8)	< 0.3-8.5 (0.9)	< 0.3-45 (8.3)	N.D.– 4.9 (0.9)	< 0.3-34 (14)	N.D.– 3.0 (0.6)	< 0.3-18 (5.9)	N.D.– 15 (2.3)	N.D.– 13 (1.8)	0.9-190 (23)
Africa	Nigeria[79]	-	-	0.01-0.25	0.01	0.02-0.42 (0.09)	0.009-0.13	0.009-0.60	0.009-0.16	0.009-0.28	0.01-0.14	0.01-0.04	0.012-0.54 (0.27)
	Kenya[21]	-	-	< 0.1-0.57	-	0.03-0.35 (0.17)	< 0.02-0.07	< 0.03-0.34	0.01-0.4 (0.1)	0.01-0.26 (0.11)	-	< 0.04-0.83	< 0.07-0.67 (0.44)

N.D. means not detected

shortcomings in analytical facilities that hinder monitoring capabilities [86]. Therefore, comprehensive monitoring and management strategies are important to mitigate PFAS contamination in sludge, particularly in regions with high levels of urbanization and industrialization.

Besides PFOS and PFOA, other PFAS significantly contribute to the total PFAS concentrations present in sludge. Fredriksson et al. [33] indicated that perfluoroundecanoic acid (PFUnA) (up to 120 ng/g dw) and PFDA (up to 69 ng/g dw) were the main contributors to the total PFAS in sludge collected at 4 WWTPs in Sweden during 2004 and 2010. This is probably attributed to their historical use in various industrial and consumer products [33]. Perfluorohexanoic acid (PFHxA) emerged as a significant contributor to the overall PFAS concentrations in sludge matrices from China, Thailand, and Switzerland, with concentrations over 90 ng/g dw (Table 1). The abundance of PFHxA in sludge matrices is associated with the use of conventional PFAS in industrial applications including firefighting foam and textiles [55]. Concentrations of PFBA (<4 ng/g dw) and PFBS (<5 ng/g dw) were lower than other PFAS in most countries shown in Table 1. Variations in the diversity and concentrations of PFAS in sludge are likely related to the alkyl chain length [106]. The dominant adsorption mechanism differs between short-chain and long-chain PFAS, with the size effect playing a crucial role for short-chain PFAS, particularly for C2-C4 [106]. Other factors such as PFAS manufacturing history, industrial application, and domestic end-use, also contribute to the variations in PFAS concentrations in sludge [66].

3.2. Source, migration pathway, and environmental risks of PFAS in sludge matrices

WWTPs are one of the most focused point sources for PFAS, where a considerable proportion of PFAS are adsorbed to sludge and are eventually released into the environment [100] (Fig. 2). PFAS in WWTPs are mostly sourced from the direct release of PFAS-containing products via industrial discharge (e.g., manufacture, and domestic products such as food packaging, cosmetics, etc.), sewage wastewater, and the in-situ formation of PFAS from PFAS precursors present in wastewater [24].

During the wastewater treatment process, microorganisms and enzymes can transform PFAS precursors into PFAS [19,57]. Fluorotelomer alcohols (FTOH), commonly found in concentrations of tens of ng/L, are typically converted into PFCA during aerobic wastewater treatment processes [19]. Additionally, other precursors, such as N-ethyl perfluorooctane sulfonamide (N-EtFOSA) and N-ethyl perfluorooctane sulfonamidoethanol (N-EtFOSE), may also be biologically transformed into PFOS [7]. The degradation of PFAS precursors contributes to increased diversity and concentrations of PFAS, such as PFCA and PFSA, in WWTPs [104]. Moreover, PFAS with increased alkyl chains are less volatile and more hydrophobic, leading to their enhanced adsorption on sludge matrices due to their stronger protein affinities in these matrices during biological wastewater treatment [2,106]. Furthermore, conventional WWTPs lack specific treatment/removal processes for treating and removing PFAS. As a result, sludge from WWTPs becomes a major sink of accumulation for both new and unremoved PFAS [2,24].

PFAS in sludge matrices can pose potential risks if released into natural environments. Sludge incineration, landfill, and agricultural reuse are the primary pathways for PFAS release from sludge [93]. Short-chain PFAS (C4-C8) can be transported via the atmosphere to soil and water matrices [1]. WWTPs are significant sources of PFAS in the air, particularly after the aeration process. PFBA concentration reached up to 116 pg/m^3, representing a 1.6-fold increase compared to levels found at primary and secondary clarifiers [1]. Hamid and Li [41] found that WWTPs sites exhibited total PFAS concentrations 1.5 to 15 times higher than locations without WWTPs emission. Notably, sludge incineration and landfill activities also contribute to PFAS air emission (Fig. 2). Total concentrations of neutral and anionic PFAS in the air of Tianjin City, China in 2016 showed 400–1121 pg/m^3 in incineration sites and 674–19262 pg/m^3 in the landfill sites. These concentrations were 2–3 times and 7–10 times higher than those found in upwind sites, respectively [92]. PFAS in the air can undergo photodegradation and/or long-distance atmospheric transport and can therefore lead to the presence of PFAS in remote locations [41]. This suggests that PFAS contamination is not solely restricted to the immediate vicinity of WWTPs and disposal sites but can be dispersed more widely, impacting ecosystems and human health in remote areas.

Additionally, during landfill processes, PFAS can migrate from sludge to landfill leachates [92]. Numerous studies have documented the occurrence of PFAS in landfills and their leachates among various

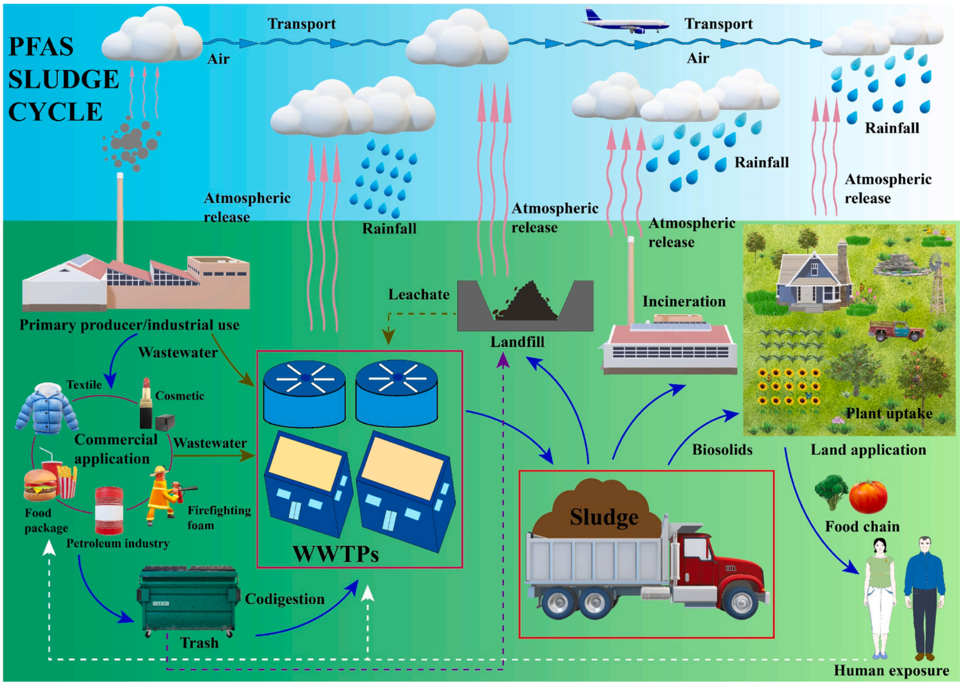

Fig. 2. Sludge-based PFAS cycle in the environment.

countries, including the USA, Australia, and China [25]. For example, leachates from Australian landfills revealed varied PFAS concentrations from 1068 to 6878 ng/L, with PFHxA being the most prevalent at 450–2700 ng/L [35]. Typically, these PFAS-containing landfill leachates are sent back to WWTPs, forming a cycle of PFAS transfer among sludge, landfills, and WWTPs (Fig. 2).

Biosolids are frequently used as fertilizers on agricultural land to enhance crop production, introducing PFAS into soil environments [46]. Some of the PFAS in the soil are taken up by plants and organisms, providing a pathway for PFAS to enter food chains. It has been reported that the total concentrations of PFAS in celery shoots, radish shoots, and pea fruit grown in biosolids-amended soil were up to 817, 279, and 236 ng/g, respectively [12]. PFAS bioaccumulation depends on the concentration and physiochemical properties of PFAS, plant species and physiology, and soil types [41]. The consumption of PFAS-contaminated crops increases the risk of exposure to PFAS for both humans and livestock, particularly the youngest children (1–2 years) [14]. The accumulation of PFAS in animals after exposure has been shown to alter their immune function, causing tumors and obesity, as well as hepatic/lipid metabolic toxicity [32]. Furthermore, human exposure to PFAS has displayed positive associations with COVID-19 occurrence, incidence, mortality, and vaccination effects during the recent COVID-19 outbreaks, indicating a substantial impact of PFAS on human immune systems [60,73].

Contamination of air, soil, surface water, and exposure of humans or livestock to PFAS can lead to the presence of PFAS in sludge and biosolids, consequently accelerating the PFAS distribution in natural environments (Fig. 2). Therefore, continued research on the occurrence and fate of PFAS in sludge is required to ensure adequate safety measures for PFAS in sludge, and ultimately to regulate their environmental distribution.

3.3. International regulations

With increasing concerns about the occurrence and exposure risks of PFAS, some countries have cautiously examined the use of PFAS-related products, and regulations have been implemented to phase out the production and use of PFAS. In July 2021, five European countries (Denmark, Germany, the Netherlands, Sweden, and Norway) announced their intention to prohibit the manufacture and use of the entire class of PFAS chemicals [31]. Australia has taken measures to prevent the release of PFAS into the environment. In 2015, the South Australian government developed an amendment that banned the use of fluorinated foams, which came into effect in 2018 [81]. The Queensland government announced to phase out firefighting foams that contain PFAS by 2019 [74]. Similarly, the New South Wales government prohibited the use of PFAS-containing firefighting foams in 2021 [30]. In the USA, Maine became the first state to implement a ban on the sale of PFAS products since 2023, and numerous other states have imposed similar restrictions for food packaging, textile articles, and cosmetics [85]. These actions reflect a growing recognition of the necessity to confront the hazards linked to PFAS and safeguard both the environment and public health.

Apart from regulations on the consumption of PFAS products, some countries have made restrictions on permissible PFAS concentrations in biosolids and their use (Table 2). Currently, most regulations are imposed for soil, whereas some countries restrict the PFAS levels in sludge/biosolids (Table 2). The regulatory values for PFAS in sludge and soil differ significantly among countries. In the United Kingdom, the guideline value for PFOA in sewage sludge disposal is 46 ng/g dw, while in Austria, it is 100 ng/g dw for the sum of PFOA and PFOS (Table 2). For agricultural application of biosolids, the permissible PFAS concentrations in Germany are below 100 ng/g dw for the sum of PFOA and PFOS. For soil matrices, the limits for PFOS are 100 ng/g dw in Norway and 390 ng/g dw in Denmark, which is much higher than the values established by the Netherlands (2.3 ng/g dw), USA (5.2 ng/g dw) and Canada (10 ng/g dw).

Table 2
International regulations towards PFAS residue in sludge, biosolids, and soil environments.

Media	Country	Contaminant	Regulation value (ng/g dw)
Soil	Queensland, Australia[27]	PFOS	1
		PFOS+PFHxS	2
		PFHxS	3
		PFOA	4
		PFBA, PFPeA, PFHxA	1
		Sum C9 -C14 PFCA	10
		PFSA	1
		n:2 FTS	4
	USA[42]	PFOS	5.2
		PFOA	2.5
		PFBS	1900
	Canada[42]	PFOS	10
	Norway[3]	PFOS	100
	The Netherlands[53]	PFOS	2.3
	Denmark[53]	PFOS	390
		PFOA	1300
		PFOSA	390
Sewage sludge	United Kingdom[26]	PFOS	46
	Austria[26]	PFOS+PFOA	100
Soil fertilizers	Germany[29]	PFOS+PFOA	100

The State EPA in Queensland, Australia, also established the first biosolids-specific guidance in Australia, where trigger values for PFAS in soil were set as 1 ng/g dw for PFOS and 4 ng/g dw for PFOA [27]. This variation may result from scientific uncertainty in risk evaluation, technical abilities, and decision-making, as well as social, political, and economic impacts from relevant stakeholders [22]. Numerous developing and least-developed countries have yet to establish regulatory values for PFAS in biosolids and soil. The limited technical capabilities and high cost of analysis impede the monitoring and regulation of PFAS contamination in biosolids in these countries [22]. To establish more effective PFAS regulatory standards and reduce PFAS pollution, international cooperation and information sharing on PFAS distribution, composition, fate, and analytical techniques are imperative.

4. Treatment approaches for PFAS in sludge

PFAS can be transported into other environmental matrices (particularly soil) during sludge treatment which results in higher PFAS concentrations than environmental concentrations [46]. With growing concern about PFAS in the environment, many treatment technologies, including physical treatments, chemical treatments, and biological treatments, have been proposed to remove PFAS from sludge matrices before entering the environment (Fig. 3). The details of these treatment approaches are discussed in the following sections.

4.1. Physical treatments

PFAS in sludge can leach downwards, leading to contaminant migration to soil, groundwater, and plants during land application and landfilling [25]. Adsorption is the main physical method for addressing PFAS in sludge (Fig. 4). Remediation of PFAS in aquatic environments through adsorption has been established in numerous studies [34], and has been recently demonstrated to be useful for removing PFAS from sludge and soil matrices [110]. The functional head groups of the PFAS molecules (e.g., -COOH and -SO_3H) are typically adsorbed by positively charged adsorbents through electrostatic binding and hydrophobic interactions [38]. PFAS adsorption is affected by the surface chemistry of both adsorbents and PFAS species [23].

Efficient adsorbents for common PFAS from sludge in laboratory, pilot, and full-scale studies include biochar, RemBind® products (a

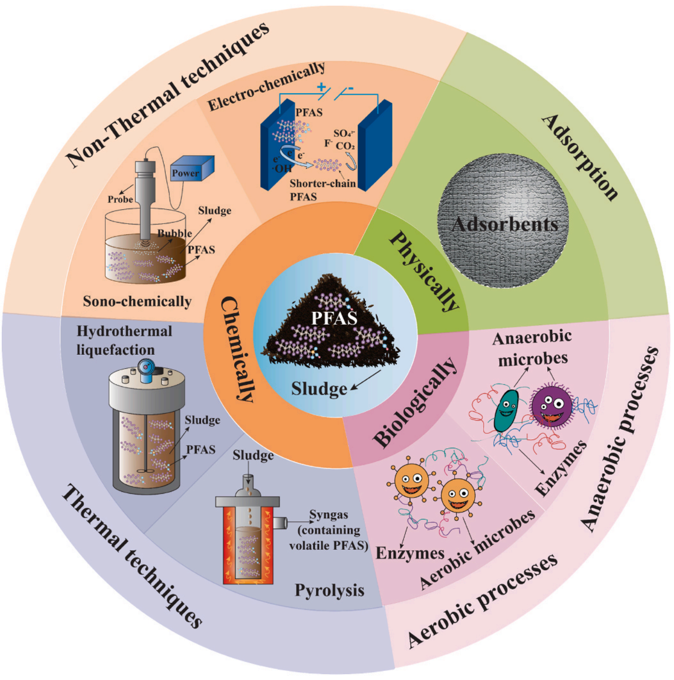

Fig. 3. Treatment approaches for PFAS remediation from sludge.

blend of amorphous aluminum hydroxide (pseudo-boehmite), kaolin clay, and activated carbon), and activated carbon (such as powdered activated carbon and granular activated carbon) with varying efficiencies. RemBind® products and activated carbon have demonstrated higher PFAS stabilization efficiencies in sewage sludge (89% for RemBind®100X, 75% for activated carbon and 51% for RemBind®100) than biochar (22%) [110]. Moreover, when added to PFAS-contaminated soil, the addition of 1% w/w of RemBind®100 resulted in an 81–87% reduction in PFAS leachability from the soil compared to unamended soil [47]. These findings underscore the potential of RemBind® products as effective adsorbents for PFAS removal.

The adsorption performance of PFAS is also significantly influenced by specific PFAS species. PFAS exhibiting higher hydrophobicity generally display greater adsorption performance, as observed in studies using various adsorbents such as activated carbon and resins [23]. For instance, PFSA are more extensively adsorbed when compared to PFCA, mainly due to their higher hydrophobicity [23]. This is reflected in the decreasing sorption affinity of PFOA (30–54 mL/g of the maximum value of sorption solid-liquid distribution coefficients ($K_{d,Max}$)), compared to PFOS (140–281 mL/g of $K_{d,Max}$) [65]. This emphasizes the significant role of -SO_3H in adsorption as compared to the -COOH group. Long-chain PFAS exhibiting the same functional groups (-SO_3H or -COOH) demonstrated greater adsorption abilities than short-chain PFAS towards adsorbents, owing to their hydrophobic properties [96]. This is evidenced by the greater $K_{d,Max}$ of PFOS than PFBS [65].

In addition to PFAS species and absorbents, adsorption performance is associated with pH, temperature, and organic matter [40,45]. For instance, anionic PFAS commonly interacts with positively charged adsorbents via electrostatic binding. Lower pH values enhance the electrostatic interaction between anionic PFAS and adsorbents, resulting in increased adsorption of PFAS to adsorbents [87]. Guo et al. [40] previously reported that an increase in pH decreased the quantity of adsorbed PFOS when corn straw biochar was used. Moreover, the impact of pH varies when certain quantities of divalent cations, such as Mg^{2+} and Ca^{2+}, are present. Increasing the pH led to the formation of additional basic sites on the adsorbent surfaces, facilitating the binding of divalent cations and hence improving PFAS adsorption from sludge via the divalent ion bridging effect [15]. Thus, maintaining an appropriate pH value is crucial for the effective remediation of PFAS from sludge or soil.

Temperature is an additional factor affecting the adsorption of PFAS on adsorbents. The adsorption capacity for PFAS on biochar increased as the temperature rose from 288 K to 308 K at pH 7.0 [40]. Furthermore, PFAS head groups typically bind to hydroxyl groups of organic matter through covalent binding, electrostatic interactions, and the hydrophobic effect [98,115]. The hydrophobic effect was found to cause interaction between the carbon chain of PFAS and organic matter [45]. These interactions create difficulties in leaching PFAS from sludge. Higher organic matter levels correspond to greater PFAS adsorption on

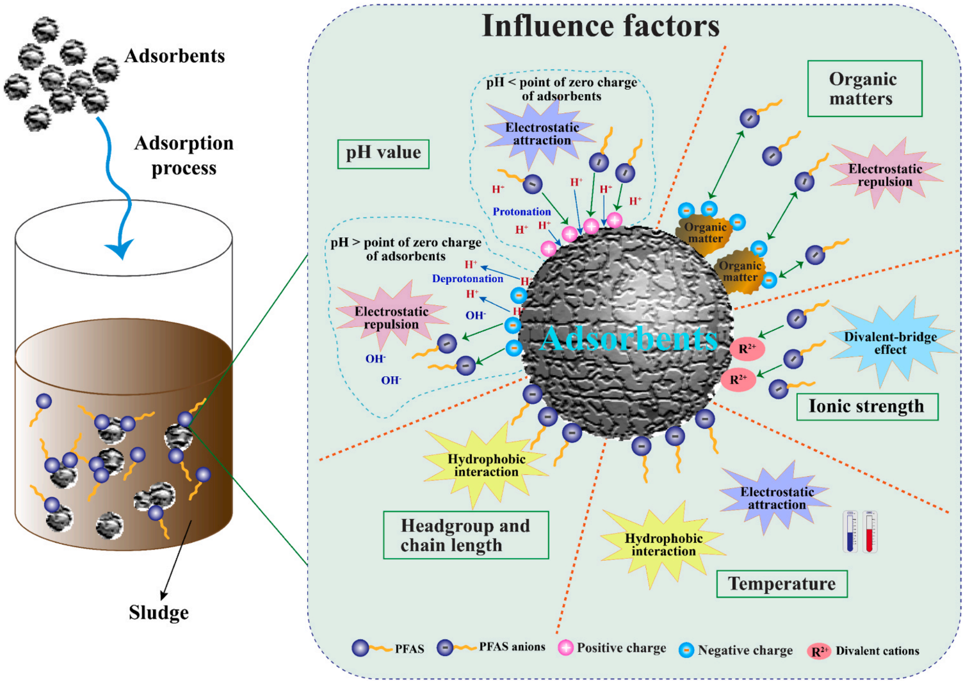

Fig. 4. Adsorption mechanism for PFAS from sludge.

sludge or soil [98], affecting PFAS remediation via adsorbents. This correction between organic matter content and PFAS adsorption provides implications for using adsorbents in PFAS remediation.

4.2. Chemical treatments

Chemical treatments of PFAS in sludge can be grouped as non-thermal (sonochemical degradation and electrochemical oxidation) and thermal (pyrolysis and hydrothermal liquefaction) approaches.

4.2.1. Non-thermal treatments

The ultrasonic method is promising for PFAS degradation due to its advantages of cleanliness, energy efficiency, and high safety [17]. Approximately 97% of PFAS in solutions was degraded during the sonochemical process, with degradation mechanisms involving pyrolytic processes and reactive radicals [97]. During sonication, microbubbles are continuously formed by the compression and rarefaction effects of the sono-waves [17]. The collapse of these bubbles can generate high pressure and temperature (approximately 5000 ℃ of temperature and 500 bar of pressure) used for PFAS pyrolysis [93]. Highly reactive radicals (such as H· and ·OH) generated during sonochemical treatments also contribute to PFAS degradation [17].

The degradation performance of ultrasonic treatment varies with multiple parameters, including initial concentration, PFAS species, temperature, and ultrasonic frequency [17,68]. PFAS degradation and defluorination initially increased with higher initial concentrations but then decreased due to saturation kinetics [75]. This was evidenced by the defluorination rates of 9.5×10^{-4}, 6.0×10^{-3} and 5.1×10^{-3} mM/min for initial PFOS concentrations of 10, 100, and 460 μM, respectively, at 1 mHz of sound frequency [75]. Similarly, at 20 ℃, 375 W of ultrasonic power, 180 pM of PFOA was the optimal initial concentrations for degradation rate and beyond, the rate decreased [72]. Chemical structures also affect PFAS degradation, with C4 PFSA degrading faster than C4 PFCA, and C4 PFAS degrading slower than C6 and C8 PFAS [16].

Furthermore, increasing temperature decreases PFAS removal efficiency, due to reduced adsorption and mass transfer rate of PFAS to the water-bubble interface, and bubble activity [17,68]. Temperature of 20 ℃ was optimum for PFOA and PFOS degradation at 375 W of ultrasonic power [72]. Ultrasonic frequency is also vital in PFAS removal, with 1 MHz being the optimal for PFAS within a 10–100 μM concentration range [75], whereas low frequencies (20 kHz) are ineffective for PFAS removal from sludge [112]. Dual-frequency ultrasounds (43 kHz for a high-frequency bath and 20 kHz for a low-frequency ultrasound probe) with persulfate can effectively address the limitations of low ultrasonic frequencies, achieving 64–71% degradation for 14 PFAS in soil [54]. Therefore, optimizing the operating conditions can provide practical options for PFAS removal using ultrasonic treatment.

Electrochemical oxidation is commonly used to treat organic compounds [39], which also shows potential for PFAS degradation by applying an electric current to the system [23], resulting in the direct degradation of PFAS on the electrode or indirect degradation through oxidizing agents produced at the electrode [68]. During the electrochemical oxidation process, C-F bonds are broken, causing the stepwise removal of CF_2 groups from PFAS parents and the formation of shorter-chain PFAS until complete defluorination [10].

Currently, most research has explored PFAS degradation in aqueous matrices [38,82], with only one study documenting the potential of electrochemical oxidation in soil, which achieved 52% degradation of PFOA and 33% of PFOS using 24 V constant voltage and 467–690 mA direct current [43]. One project on the electrochemical extraction and remediation of PFAS in soil was initiated in the USA on June 1, 2020 [89]. Therefore, further research is required to examine the effects of electrochemical oxidation and its operating conditions on PFAS removal efficiency in sludge and soil matrices due to their distinct physicochemical properties compared to water matrices.

4.2.2. Thermal treatments

Thermal treatments such as pyrolysis (300 to 1000 ℃ in oxygen-free environments) and gasification (800–1650 ℃ with low oxygen) have shown promising prospects for PFAS remediation in sludge and soil [10]. These processes involve the cleavage of C-C and C-F bonds in PFAS, leading to their transformation into gas-phase compounds and subsequently conversion into HF and/or fluoride organics [10,107].

Factors such as PFAS structures, temperature, and catalyst, affect PFAS removal efficiency by pyrolysis. Over 96% of PFOA and PFOS were removed, compared to $< 75\%$ for PFBS, PFPeA, and PFHpA, suggesting the significance of chemical structures of PFAS during pyrolysis. The low removal probably due to the formation of these compounds during the process [50]. Additionally, increasing the temperature during pyrolysis significantly decreased the concentrations of PFAS by-products in biochar and pyro-gas. For instance, raising the temperature from 500 ℃ to 700 ℃ resulted in an 83.7% decrease in PFAS emissions in pyro-gas and an 84.3% increase in the overall PFAS removal from biosolids [67]. Bamdad et al. [8] also found that increasing temperature from 500 ℃ to 700 ℃ improved PFAS removal in biosolids to 99.6%. Some minerals and metals present in biosolids, such as Ca, Fe, Zn, and Cu, can act as catalysts for PFAS destruction, resulting in lower energy and temperature requirements [50]. Semi-pilot or pilot research has demonstrated over 99% elimination of PFAS using pyrolysis [50,84], indicating its real-world practicality.

Hydrothermal liquefaction is another technology for PFAS remediation in sludge, which involves the conversion of organic matters in sludge into liquid biocrude oil at high temperatures (250–350 ℃) and pressure (10–25 MPa). Factors including PFAS species, temperature, maintenance time, and additives, affect PFAS degradation during hydrothermal liquefaction. PFAS with-COOH groups (PFOA, 7:3 fluorotelomer carboxylate (7:3 FTCA), and 8:2 fluorotelomer unsaturated carboxylic acid, 8:2 FTUCA) underwent $> 99\%$ decomposition at 350 ℃ for 1.5 h while -SO_3H groups (PFOS and 8:2 fluorotelomer sulfonate (8:2 FTS)) experienced limited degradation (34% and 67%, respectively) [103]. Zhang and Liang [111] also documented complete degradations for PFHpA, PFHxA, and PFOA, but limited degradation for PFOS and PFBS in sludge at 250 ℃ for 2 h. These findings demonstrate the structure-specificity of hydrothermal liquefaction for PFAS degradation.

It is necessary to maintain higher temperatures of 300 ℃ and 350 ℃ for the degradation of PFBS and PFOS, respectively, in hydrothermal liquefaction systems [103,111]. Additionally, concentrations of PFAS precursors in sludge increased initially at 300 ℃ for 0.5 h due to their release from bound and non-extractable fractions, but then the degradation efficiencies reached 50% as treatment time increased to 2 h [111]. The addition of $Ca(OH)_2$ further enhanced the elimination of PFAS precursors [111]. These indicate the significance of the operational conditions and additives in converting PFAS precursors to PFCA during the hydrothermal liquefaction process, which warrants further research.

4.3. Biological treatment

Some past work has examined the transformation and fate of PFAS based on biological processing, providing intriguing insights. This section comprehensively discusses the removal and mechanism of PFAS under aerobic and anaerobic conditions based on the early and newly published studies (Fig. 5).

4.3.1. Aerobic condition

Aerobic conditions facilitate the biotransformation of PFAS embedded in sludge and soil, ultimately leading to their degradation [18,61]. Under such conditions, PFAS precursors, such as FTOH, fluorotelomer phosphate diester (diPAP), and N-EtFOSA), usually undergo conversion into PFCA and PFSA due to their short half-lives (Table 3). For instance, 6:2 diPAP was biotransformed into 5:3 acid, PFPeA, and PFHxA, while 8:2 diPAP was converted into PFOA [61]. PFOA and PFOS

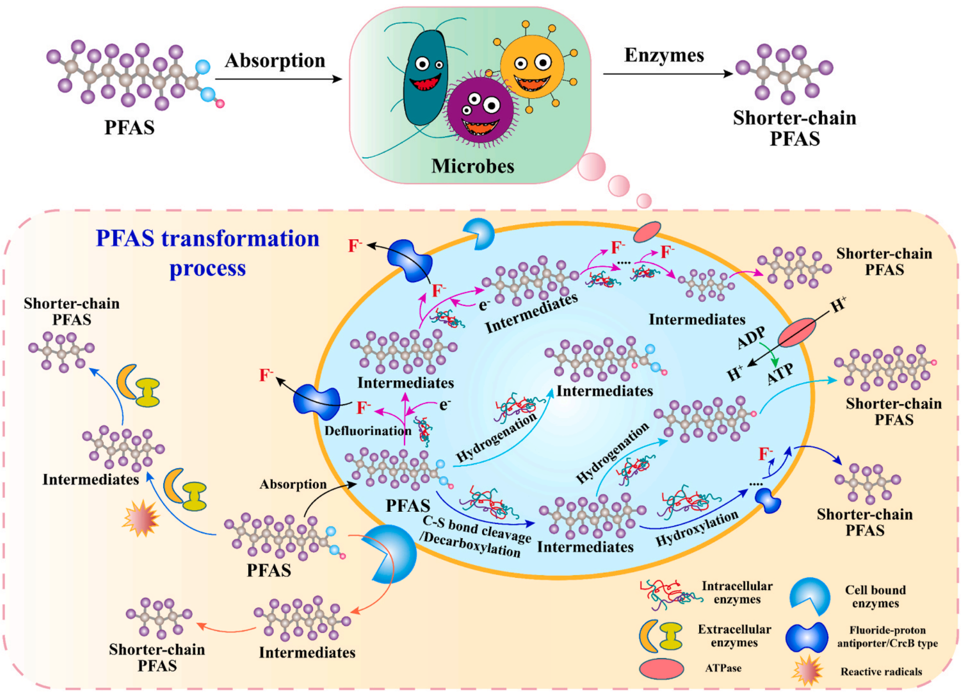

Fig. 5. Biological process for PFAS biotransformation in sludge.

in sludge can also be degraded aerobically (Table 3). Yi et al. [102] reported that approximately 32% of PFOA (initial concentration of 500 mg/L) was degraded aerobically within 72 h at 30 ℃ and pH of 7. About 67% decomposition of 1400–1800 μg/L PFOS was also observed within 48 h, leading to the formation of various shorter-chain PFSA, predominantly PFBS and PFHxS (4.0–26.0 ng/L), as well as unknown by-products due to the partial cleavage of C-C bonds in the PFOS molecule [51]. Free F^- were also released at room temperature due to the biotransformation of PFOS [20]. These findings emphasize the potential of PFAS degradation in aerobic environments.

Shorter-chain PFAS can also be aerobically degraded. Che et al. [18] identified four PFCA with anaerobic defluorination exceeding 20% among 14 commercial PFCA (C3-C5), including 3,3,3-trifluoropropionic acid (84%), 4,5,5-trifluoropent-4-enoic acid (70%), 5,5,5-trifluoropentanoic acid (37%), and 2-fluoropropionic acid (20%). The study revealed structure-dependent relationships in defluorination, emphasizing the role of specific functional groups like C-H bonds at the α-position and sp2 C-F bonds at the β-position. Unstable intermediates containing fluoroalcohol moieties were formed during the reaction, resulting in the spontaneous cleavage of C-F bonds [18]. In aerobic conditions, unsaturated PFAS can also be removed and defluorinated. For instance, 2-(Trifluoromethyl)acrylic acid was completely removed within 1 day and defluorinated completely within 2 weeks [105]. The elimination mechanism of which began with hydrogenation and resulted in the generation of 2-methyl-trifluoropropionate. Aerobically remediation of 4,4,4-Trifluoro-3-(trifluoromethyl)crotonic acid led to 50% removal and 82% defluorination degree [105]. These findings demonstrate that aerobic biotransformation of PFAS and their precursors appears promising and achievable.

4.3.2. Anaerobic condition

Anaerobic digestion is a widely used process for stabilizing sludge [94, 95] and can also lead to PFAS degradation (Table 3). Xie et al. [99] reported 10–28% reduction in PFOA levels in an anaerobic digestion system. Lakshminarasimman et al. [52] also reported < 40% removal of 7 PFAS (e.g., PFHxA, PFOA, PFDA, PFUnA, PFDoA, PFOS, PFOSA) and 71% removal for PFBS in 5 Canadian WWTPs. Several studies have focused on the biotransformation of PFAS precursors in anaerobic conditions. For example, 8:2 FTOH underwent oxidation in activated sludge to produce 2 H-perfluoro-2-decenoic acid (8:2 FTUA) and 8:2 fluorotelomer carboxylate (8:2 FTCA), which can undergo further anaerobic degradation to produce PFOA and shorter-chain PFCA (C<8) and release F^- [57]. However, Zhang et al. [109] reported that FTOHs were not the primary PFCA source in anaerobic matrices, and only a small amount of PFCA was produced in methanogenic conditions due to the absence of key enzymes for decarboxylation. Therefore, further research is required to optimize the anaerobic processes for the effective PFAS removal.

In addition to conventional PFAS species, novel PFAS alternatives can also undergo degradation in anaerobic conditions. Yu et al. [105] investigated the bio-defluorination and biotransformation pathways of unsaturated PFCA (4-(Trifluoromethyl)hexafluoropent-2-enoic acid and 4,5,5,5-tetrafluoro4-(trifluoromethyl)– 2-pentenoic acid) in anaerobic conditions. They found that reductive defluorination (-F+H) and hydrogenation (+2 H) are the two main pathways, and the priority of these two pathways was related to the number of fluorine substitutions on unsaturated carbons. These findings provide valuable insights into the potential for the transformation of PFAS alternatives in anaerobic environments.

4.3.3. Biodegradation mechanism of PFAS during biological processes

Earlier works have demonstrated that the reduction of PFAS concentration and the corresponding release of F^- during aerobic and anaerobic biodegradation are primarily dependent on microbial species and enzymes (Fig. 5). Microbial species identified in biological processes include *Acidimicrobium* sp. Strain A6 [44], *Pseudomonas* species (*P. parafulva*, and *P. plecoglossicida* 2.4-D) [102,104], and *Rhodococcus jostii* RHA1 [101]. For example, *Acidimicrobium* sp. Strain A6, an autotrophic microbe capable of NH_4^+ oxidation and Fe(III) reduction, has demonstrated the ability to degrade PFOA and PFOS in sludge. Huang and Jaffe [44] observed a 50% reduction in PFOA and the generation of four intermediates (PFBA, PFPeA, PFHxA, and PFHpA) in the A6 enrichment culture, showing higher efficiency compared to the pure A6 culture (33% reduction). Proteobacteria also significantly contribute to FTOH biotransformation [104].

In the degradation of PFAS in sludge and soil, microbial excretion of intracellular and extracellular enzymes, including cytochrome P450

Table 3
Main findings towards PFAS remediation via the biological process.

PFAS	Conditions	PFAS initial concentration	Microbial strains	Biodegradation and defluorination efficiency	Biotransformation intermediates
PFOA ($C_8HF_{15}O_2$)[102]	Aerobic, pH 7, 30 ℃	500 mg/L	*Pseudomonas parafulva*	32% degradation within 72 h; 48% degradation with 96 h with the addition of 1 g/L glucose	-
PFOS ($C_8HF_{17}O_3S$)[51]	Aerobic, pH 7-9, 30 ℃	1400-1800 µg/L	*Pseudomonas aeruginosa* strain HJ4	67% decomposition after 48 h	PFBS, PFHxS, and several unknown products
PFOS ($C_8HF_{17}O_3S$)[20]	Aerobic, 28 ℃	-	*Pseudomonas plecoglossicida 2.4-D*	75% degradation with the release of fluorine ions	PFHpA
MeU-C4c ($C_4H_3F_3O_2$) [105]	Aerobic	75 µM	dominated by *Dehalococcoides*	100% aerobic removal in 1 day and 100% defluorination within 2 weeks	Probably in CoA forms, and difficult to extract and detect
MeU-C5d ($C_5H_2F_6O_2$) [105]	Aerobic	75 µM	dominated by *Dehalococcoides*	50% removal and 82% defluorination	Hydrogenation product (MeUC5d_TP209, $C_5H_4F_6O_2$) and two defluorination intermediates (MeUC5d_TP121 and MeU-C5d_TP139)
6:2 FTUCA ($C_8H_2F_{12}O_2$) [105]	Aerobic	75 µM	dominated by *Dehalococcoides*	10% defluorination	PFHxA, PFPeA, 2 H-PFHpA, and 2 H-PFHxA
3,3,3-trifluoropropionic acid ($C_3H_3F_3O_2$)[18]	Aerobic	50 µM	activated sludge community	100% removal and 85% defluorination degree	No intermediates were detected
2-fluoropropionic acid ($C_3H_5FO_2$)[18]	Aerobic	50 µM	activated sludge community	100% removal and 21% defluorination degree after 72 h	volatile fluorinated alkane
5,5,5-trifluoropentanoicAcid ($C_5H_7F_3O_2$)[18]	Aerobic	50 µM	activated sludge community	100% removal and 37% defluorination degree within 3 h	not reported
4,5,5-trifluoropent-4-enoic acid ($C_5H_5F_3O_2$) [18]	Aerobic	50 µM	activated sludge community	30% removal and 71% defluorination degree	monofluoromalonyl-CoA
6:2 diPAP ($C_{16}H_9F_{26}O_4P$) [61]	Aerobic, 22 ℃	4.22 nmol/g soil	Soil microbes	-	5:2 sFTOH (< 2.8%), 6:2 FTOH (<2.8%), PFBA (0.73%), PFPeA (6.4%), PFHxA (6.0%), 5:3 Acid (9.3%)
8:2 diPAP ($C_{20}H_9F_{34}O_4P$) [61]	Aerobic, 22 ℃	3.37 nmol/g soil	Soil microbes	-	PFOA (2.1%), PFHxA (0.34%), 7:3 Acid (0.29%), PFHpA (0.25%)
6:2 FTOH ($C_8H_5F_{13}O$) [104]	Aerobic	0.455 mg/L	*Chlorobi*, *Proteobacteria*, *Bacteroidetes*, *Chloroflexi*, *Crenarchaeota*, *Planctomycetes*, *Acidobacteria*	-	PFBA, PFPeA, PFHxA

(*continued on next page*)

Table 3 (*continued*)

PFAS	Conditions	PFAS initial concentration	Microbial strains	Biodegradation and defluorination efficiency	Biotransformation intermediates
8:2 FTOH ($C_{10}H_5F_{17}O$) [104]	Aerobic	12.1 mg/L	*Chlorobi, Proteobacteria, Bacteroidetes, Chloroflexi, Crenarchaeota, Planctomycetes, Acidobacteria*	-	PFBA, PFPeA, PFHpA, PFHpA, PFOA, PFNA
6:2 FTS ($C_8H_5F_{13}O_3S$) [101]	Aerobic	20 mg/L	*Rhodococcus jostii* RHA1	> 99% degradation with ethanol in S-free medium	6:2 FTUCA, α-OH 5:3 FTCA, and PFHpA
PFOA ($C_8HF_{15}O_2$)[44]	Anaerobic, pH 4.5-5.0	100 mg/L	*Acidimicrobium* sp. Strain A6	60% removal in A6 enrichment cultures	PFBA, PFPeA, PFHxA, PFHpA
PFOS ($C_8HF_{17}O_3S$)[44]	Anaerobic, pH 4.5-5.0	100 mg/L	*Acidimicrobium* sp. Strain A6	47% removal in A6 enrichment cultures	PFBA, PFBS
PFMeUPA ($C_6HF_9O_2$) [105]	Anaerobic	75 µM	dominated by *Dehalococcoides*	100% biotrasformation and 10% defluorination (> 52 µM fluorine ions release)	Not reported
MeU-C4c ($C_4H_3F_3O_2$) [105]	Anaerobic	75 µM	dominated by *Dehalococcoides*	100% abiotic transformation and 10% defluorination	MeU-C4c_TP260 ($C_7H_{10}F_3NO_4S$)
MeU-C5d ($C_5H_2F_6O_2$) [105]	Anaerobic	75 µM	dominated by *Dehalococcoides*	100% abiotic transformation and 78% defluorination	Hydrogenation product (MeUC5d_TP209, $C_5H_4F_6O_2$)
6:2 FTOH ($C_8H_5F_{13}O$) [109]	Anaerobic, 35 °C	1.6 mg/L	Methanogenic microbes	-	6:2 FTCA (44 mol%), 5:2 Acid (23 mol%), 6:2 FTUCA (2-8 mol%), 5:2 sFTOH (2.5 mol%), PFHxA (0.2 mol%)
8:2 FTOH ($C_{10}H_5F_{17}O$) [109]	Anaerobic,35 °C	182 µg/L	Methanogenic microbes	-	7:3 Acid (27 mol%), 8:2 FTCA (23 mol%), PFOA (0.3 mol%)

“-” means not reported.

enzymes, alkane monooxygenases, haloacid dehalogenases, and laccase, are found to be dominant [101]. Haloacid dehalogenases and fluoroacetate dehalogenases are responsible for hydrolytic defluorination through catalyzing defluorination of mono- (MFA) di-(DFA) and trifluoroacetates (TFAs), which has been identified in the defluorination of PFAS and their precursors [18,101]. Taking 2-fluoropropionic acid as an example, it underwent hydrolytic defluorination induced by 2-haloacid dehalogenases in aerobic microbes [18]. Moreover, cytochrome P450 enzymes and alkane monooxygenase also participate in PFAS fluorination [11,101]. However, the enzymatic defluorination process may be hindered by the -SO_3H group in PFSA, even with high levels of expressed enzymes (such as cytochrome P450, alkane monooxygenases, and haloacid dehalogenases) in aerobic or anaerobic systems [101]. This suggests that desulfonation processes are required before defluorination. For the PFSA desulfonation, this process is initiated by alkanesulfonate monooxygenases (SsuD), which is an FMNH2-dependent monooxygenase and responsible for breaking the carbon-sulfur bond in different alkanesulfonates [11,101].

PFAS degradation by microbes is generally affected by pH, temperature, oxygen, and environmental matrices [11]. Temperature and pH often affect the biodegradability of PFAS through enzymatic activity during enzymatic reactions [11]. Faster transformation of EtFOSE was observed at 25 ℃ (half-life ($t_{1/2}$)= 44 days) than at 4 ℃ ($t_{1/2}$ =160 days) [9]. The biodegradation of EtFOSE was also found to be 4–5 times higher in alkaline soil (pH 7.8) than in acidic soil (pH 5.5) [108]. Oxygen is vital for PFAS biotransformation in sludge via aerobic or anaerobic processes [105]. Unsaturated PFAS showed improved removal and defluorination in aerobic conditions than in anaerobic conditions [105]. These findings highlight the importance of process conditions for effective PFAS biotransformation.

Additionally, the physicochemical characteristics of environmental matrices have significant impacts on organic matter, microbial structures, and population density, influencing the biodegradability of PFAS [11,44]. Sludge and soil offer sufficient organic matter as growth substrates for microbial consumption during the PFAS defluorination process [11]. Additional carbon sources, such as 1 g/L glucose, promoted PFOA degradability by 16% enhancement [102]. Under the same conditions, *Pseudomonas fluorescens* DSM 8341 was able to transform 6:2 FTOH into PFBA, while *Mycobacterium vaccae* JOB5 did not demonstrate that capability [48]. Therefore, it is crucial to understand the different factors that affect PFAS biodegradation by microbes to design effective remediation strategies for contaminated environments. Further research in this area can aid in identifying novel approaches and techniques for improving the biodegradation of PFAS, thereby promoting sustainable environmental management.

4.4. Limitations and prospect of different treatments

Physical and chemical methods are effective in removing and remediating PFAS and their precursors (Fig. 6). Nevertheless, these techniques are not universally effective for all PFAS classes [28]. Moreover, high cost and energy demand are accompanied by the treatment process. Furthermore, physical technology (adsorption) has rarely been observed to destroy PFAS, resulting in the accumulation of PFAS in adsorbents and creating an additional issue for disposing of PFAS-contaminated adsorbents or materials, which is also costly and challenging [28]. Therefore, focusing on the regeneration and reusability of adsorbents is crucial. Future research should aim to develop effective techniques for desorbing and recovering PFAS from adsorbents, enabling their potential reuse or proper disposal.

Although non-thermal techniques show promise in treating PFAS, limited semi-pilot and pilot research has been reported, possibly due to the complicated matrices. Therefore, practical applications require to address several issues: 1) the complexity and diversity of physicochemical properties of sludge and soil matrices in real-world environments; 2) further optimization of operation conditions, identification of degradation intermediated, and minimizing the formation of problematic byproducts; 3) further exploration of parameters such as reactor geometry and location, and energy consumption, to enhance the efficiency of PFAS degradation and practicality of these technologies. The thermal approach is more commonly applied in laboratory and pilot scale studies, achieving both sludge reduction and PFAS remediation. However, barriers to sampling emissions and measuring PFAS airborne emissions after thermal processes also need to be resolved.

Given the environmental toxicity and persistence of short-chain PFAS, it is preferable to completely degrade PFAS pollutants in sludge, biosolids, and soil matrices into F^- and carbon dioxide. Prospective techniques for remediating PFAS and their precursors involve biological approaches employing microbes under aerobic and/or anaerobic conditions. These processes simultaneously address problems of energy requirement and high cost, along with the defluorination needed for sludge/soil. The varied microbial communities naturally occurring in the sludge or soil possess the capability to induce and catalyze the biodegradation and bio-defluorination of PFAS, effectively mitigating

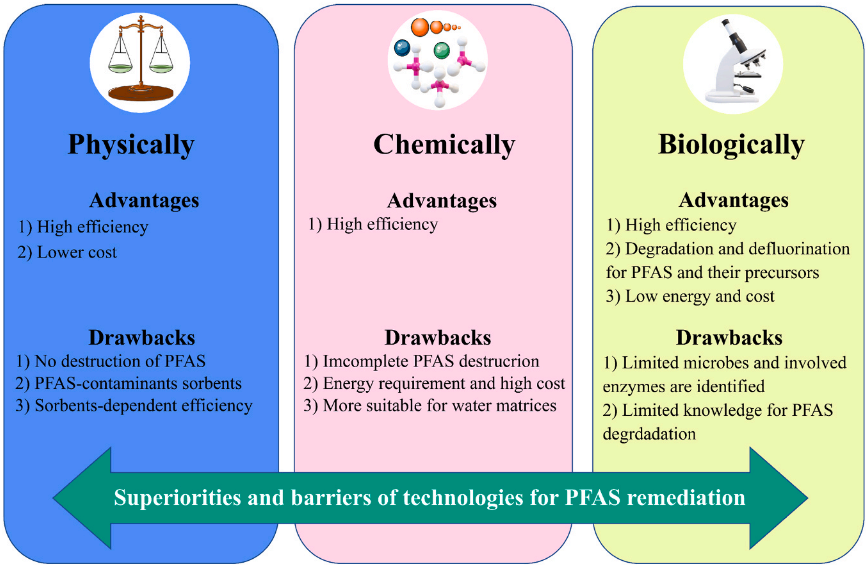

Fig. 6. Superiorities and barriers of technologies for PFAS remediation.

the generation of secondary pollutants [105,44,57].

Nevertheless, only a limited number of microbes and involved enzymes have been discovered for PFAS biodegradation, impeding the progress of biological strategies for PFAS bioremediation at the pilot scales. Further studies are required to identify microbes and enzymes linked to PFAS degradation, which can be applied through bioaugmentation to improve biodegradation and bio-defluorination efficiency. Additionally, fluorine-containing alternative products are emerging in response to the hazards of PFAS, and the feasibility of their elimination through biological methods warrants exploration. It is valuable to identify the degradation pathway of PFAS using advanced analytical technology, particularly intermediates, and to conduct risk assessments before full-scale pilot implementation.

5. Conclusions

The widespread utilization of products containing fluorine has led to the emergence of PFAS in the environment and posed environmental and health hazards. This review presents recent research on the global distribution, origins, and worldwide regulations of PFAS in sludge (and biosolids). PFAS in sludge generally come from point (e.g., industrial factories, WWTPs, landfills, and incineration sites) and non-point (e.g., rainfall and runoff) sources. The PFAS concentrations in sludge matrices across the world are up to thousands of ng/g dw, while it has poorly investigated in most developing and least-developed countries. This is due to various factors including social, political, and economic influences. Analytical skills including sample preservation, preparation, extraction, and instrument analysis are discussed in detail. Extraction is crucial in determining sludge samples, which differ from regular water samples. PFAS detection commonly utilized non-targeted screening technologies. Due to the toxicity and persistence of PFAS in sludge environments, this study discusses cutting-edge technologies for PFAS removal and defluorination, including factors that influence their efficiency, advantages, and limitations. Despite the proposed regulations and remediation techniques towards PFAS, it is joint efforts of scientists, policymakers, and industrial producers that will remain crucial in addressing the issue related to the PFAS residue problems in the sludge environments.

CRediT authorship contribution statement

Zhou Ting: Writing – Original draft, Visualization, Methodology, Investigation, Data curation, Conceptualization. **Li Xuan:** Writing – review & editing, Methodology, Conceptualization. **Liu Huan:** Methodology, Writing – review & editing. **Dong Shiman:** Software, Methodology, Data curation. **Zhang Zehao:** Writing – review & editing. **Wang Zhenyao:** Methodology. **Li Jibin:** Writing – review & editing. **Nghiem Long D.:** Writing – review & editing. **Khan Stuart J.:** Writing – review & editing, Methodology. **Wang Qilin:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization.

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.

Data Availability

Data will be made available on request.

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