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# A review of per- and polyfluoroalkyl substances in biosolids: geographical distribution and regulations

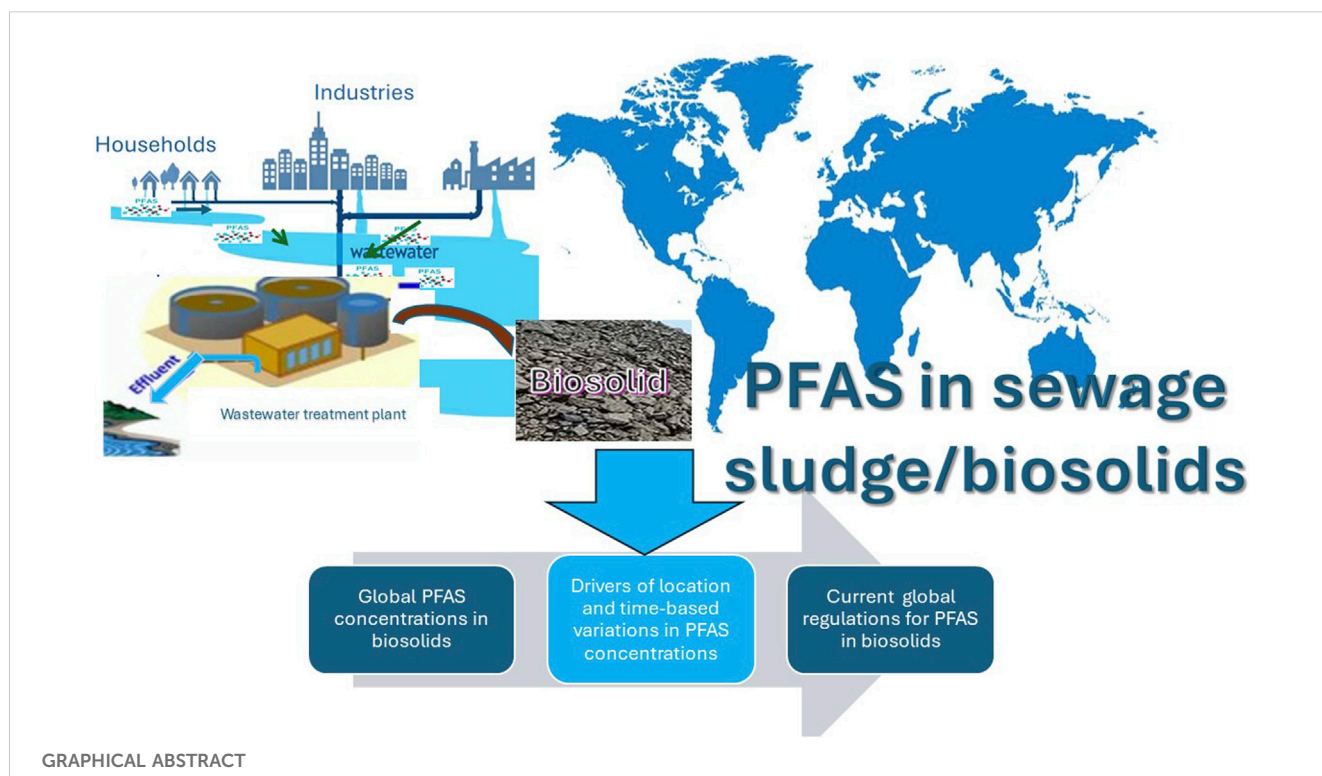
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Applying sewage sludge and biosolids to agricultural lands has become an increasingly essential aspect of sustainable waste management and circular economy as it contributes positively to nutrient recycling, soil fertility and environmental health. Due to the widespread presence of per and polyfluoroalkyl substances (PFAS) globally, wastewater treatment plants have become a sink for PFAS. PFAS resist degradation by conventional wastewater treatment processes and are usually adsorbed to sewage sludge and biosolids. However, there have been significant concerns that land application of sewage sludge and biosolids could become a probable pathway for PFAS to enter the food chain. This article assessed the global sewage sludge/biosolids generation and country-to-country management methods through a systematic review. The global occurrence, distribution and prevalence of different classes of PFAS were assessed. We also evaluate the factors influencing PFAS contamination in sewage sludge/biosolids and the existing regulations on the upper limit of PFAS in biosolids before their disposal or application to farmland (or other usages). Additionally, most reports revealed high PFAS concentrations in influent, effluent, sewage sludge and biosolids generated worldwide. Overall, recorded PFAS concentration on a global scale varied from 2.2 to 2,156 ng/L (influent), 1.9–4,800 ng/L (effluents) and 2.1–500,000 ng/g (biosolids). While most studies focused on legacy PFAS detection, recent studies have revealed the prevalence of diPAPs in high concentrations in sewage sludge and biosolids, contributing from 40% to 95% of the total PFAS concentration. Across all PFAS classes, PFAAs and diPAPs were the dominant groups exhibiting elevated detection rates (35%–95%). Due to documented PFAS contamination in agricultural lands, rigorous regulations need to be instituted to govern the application of these biowastes on agricultural lands. However, several countries lack data on the level of PFAS in the sewage sludges they generate, and there are currently few or no regulations guiding their application to farmlands. Notably, the diPAPs class of PFAS was shown to be present in biosolids and sewage sludge; their inclusion in the list of PFAS required in standardized analytical methods and risk assessment becomes imperative.

## KEYWORDS

sewage sludge, biosolids, nutrient recycling, PFAS, agricultural land, wastewater



## 1 Introduction

Sewage sludge is a slurry residue produced from wastewater treatment operations. Typically, sewage sludge comprises approximately 3% solids and 97% liquids (Kumar et al., 2017). On the other hand, biosolids are treated sewage sludge that adheres to specific standards for its utilization or disposal, with an approximate solid content of nearly 90% (USEPA, 2021). Different methods, from composting, acidic oxidation/disinfection, thermal drying, alkaline stabilization, and aerobic and anaerobic digestion, are used for biosolids treatment. These treatments distinguish the physicochemical properties of biosolids from those of sewage sludge (Mohajerani et al., 2017).

The concept of resource recovery from wastes has provided an alternative use for biosolids, sewage sludge and composts because these wastes are rich in organic matter and essential plant nutrients, such as phosphorus, nitrogen, potassium, magnesium, calcium, and sulphur (Dad et al., 2019). The total nitrogen, phosphorus, and potassium concentrations in biosolids can be as high as 3%, 1.5%, and 0.7%, respectively, while the organic content can be from 30% to 40% of organic matter (Kumar et al., 2017). However, the methods used to stabilize sewage sludge greatly influence the contents of organic matter and nutrients and the fertilizer value of the resulting biosolids (Dad et al., 2019). Owing to the chemical composition inherent in these biowaste materials, they find application as fertilizers in agricultural practices, land reclamation and reforestation efforts. This utilization is a viable alternative source of essential macronutrients for soil enrichment (Rigby et al., 2016; Dad et al., 2019). Studies have also shown that biosolids-based soil amendments enhance plant accessibility of nitrates, ammonium ions, and nitrites in the soil as a consequence of the

mineralization of organic nitrogen (Sharma et al., 2017; Wang et al., 2017).

Applying sewage sludge/biosolids and composts to agricultural lands has become an increasingly essential aspect of sustainable nutrient management as it contributes positively to nutrient recycling, soil fertility and properties. There are clear circular economy arguments for closing the nutrient and carbon loops so that nutrients extracted from the soils to produce our food are returned to the soils to maintain their fertility and quality. The use of nutrients from biosolids also replaces chemical fertilizers that would otherwise need to be produced, mined and transported with all of the associated environmental impacts.

Although the land application of biosolids is advancing in popularity and is beneficial in terms of improving soil health and resource recovery, there are recent apprehensions concerning environmental and human health hazards associated with their application to agricultural land. Several organic and inorganic pollutants have been found in biosolids that can potentially be transferred from the applied biosolids to crops (Clarke and Cummins, 2015; Gworek et al., 2021; Hušek et al., 2022; Kumar et al., 2022; Thompson et al., 2022; Adu et al., 2023; Marchuk et al., 2023). Recent studies have called into question the agricultural use of biosolids due to the documented reciprocal interaction between pollutants such as trace metals, per- and polyfluoroalkyl substances (PFAS), pharmaceuticals, hormones, polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), pesticides and plasticizers, polycyclic aromatic hydrocarbons (PAHs), flame suppressants, alkyl phenols, as well as other contaminants that could be potentially released in environment (Rathankumar et al., 2020). Many of those contaminants pose a significant risk because they can produce direct biological effects such as genotoxicity,

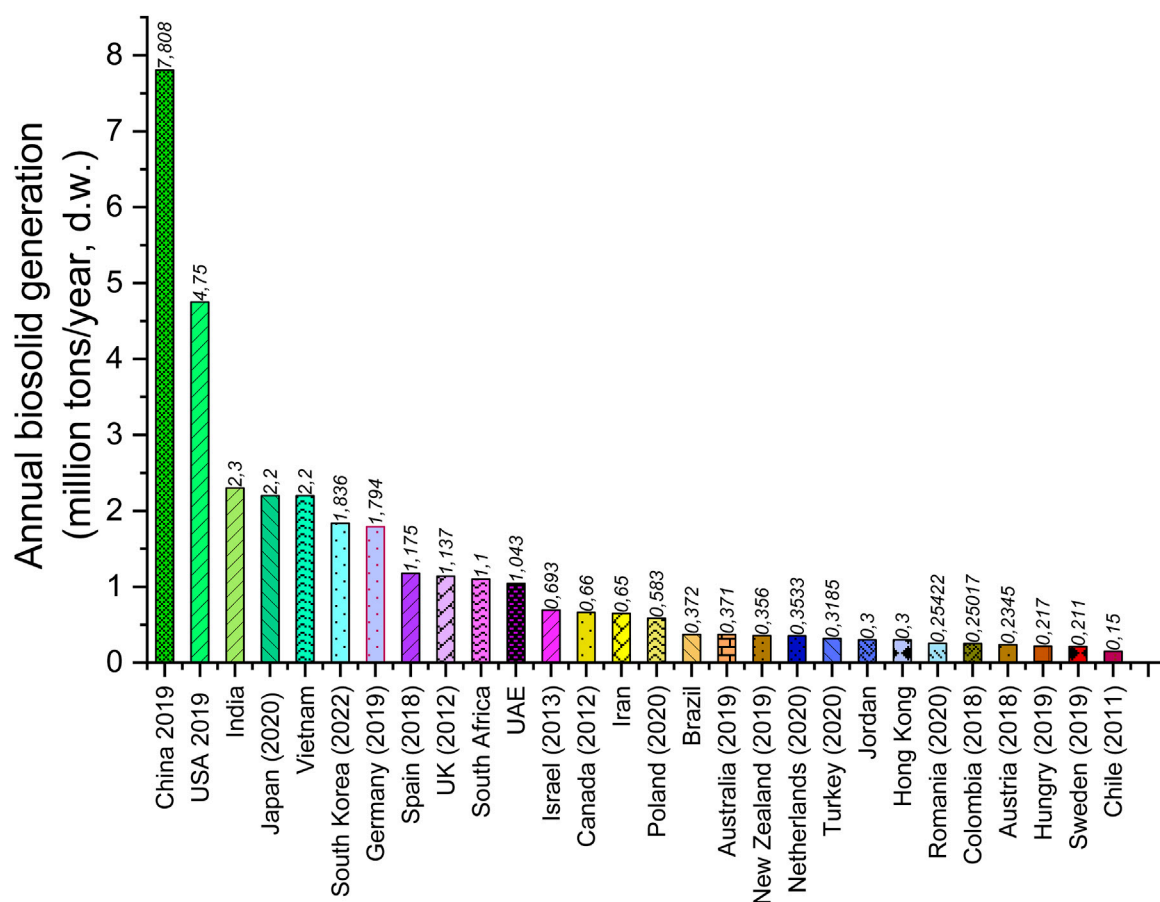


FIGURE 1

Annual generation of biosolids (BS) by selected countries. Source [China (Wei et al., 2020), United States (USEPA, 2021), Japan (Yosuke Matsumiya, 2020), Spain, Germany, United Kingdom, Poland, Netherlands, Türkiye, Romania, Austria, Sweden, (Eurostat, 2022), Canada (Canadian Council of Ministers of the Environment, 2012), Iran, South Korea, Jordan, Brazil (UN-Habitat, 2008; The Australian and New Zealand Biosolids Partnership ANZBP, 2019) New Zealand and Australia, Colombia (Venegas et al., 2021), Chile and Israel (Organisation for Economic Co-Operation and Development OECD, 2017), India, South Africa, Hong Kong (Sharma et al., 2017)].

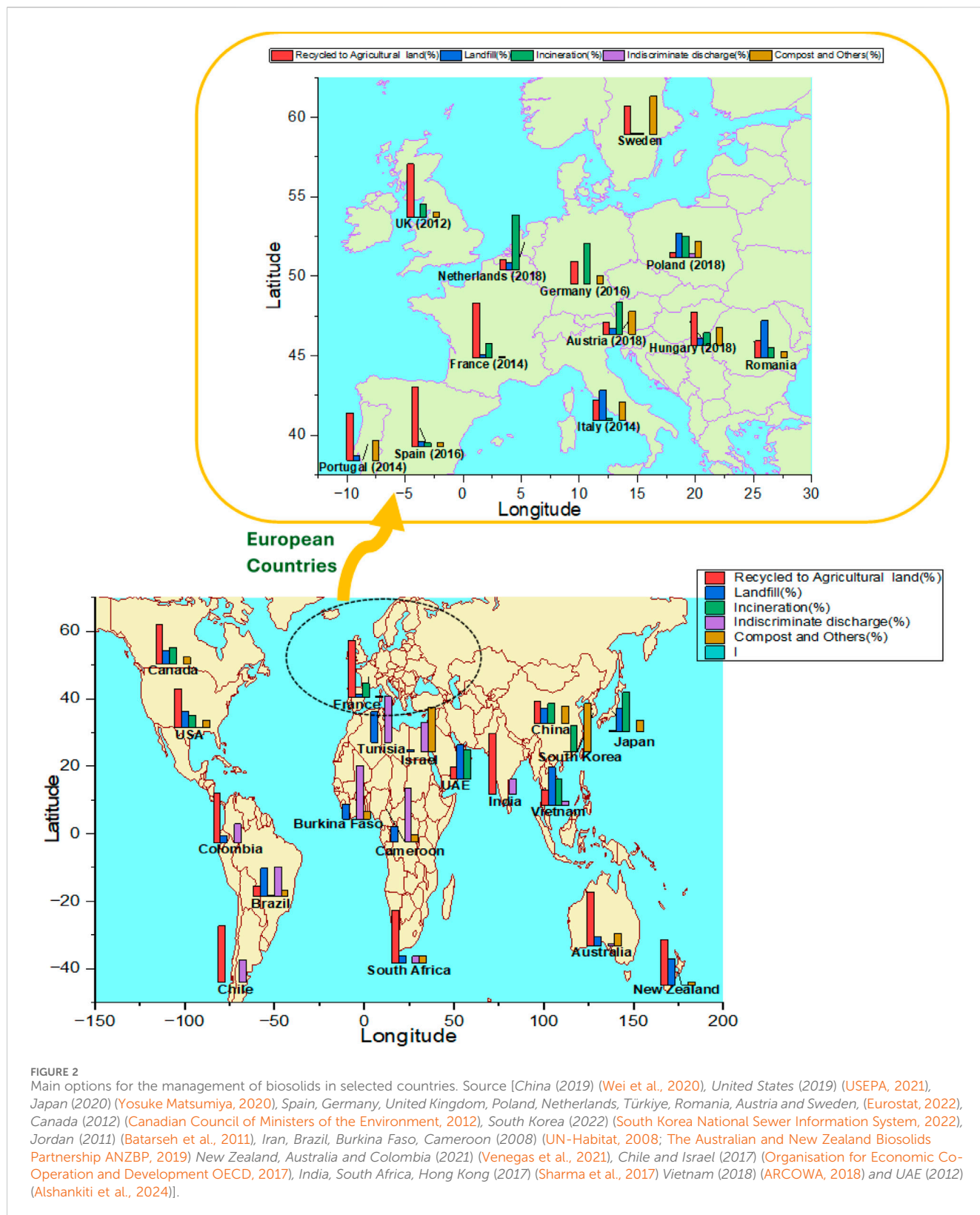
endocrine disruption, neurotoxicity, immunotoxicity or reproductive defects in living organisms (Mohajerani et al., 2017; Rathankumar et al., 2020). Irrespective of how sewage sludge/biosolids are treated, organic and inorganic contaminants are found in biosolids; however, substantial treatment processes will usually significantly abate the volatile components and pathogens (Mohajerani et al., 2017).

PFAS are a class of synthetic aliphatic compounds characterized by a completely fluorinated carbon chain (per-) or partially fluorinated carbon chain (poly-) making up the hydrophobic tail of the compound. This hydrophobic tail is usually connected to at least one hydrophilic head that constitutes the functional group of the compound. PFAS have unique chemical features due to their extremely polar and strong carbon-fluorine linkages, such as excellent thermal and chemical stability and exceptional surface-tension-lowering capabilities (Xiao, 2017). Because of their hydrophobic and hydrophilic characteristics, PFAS have been extensively utilized globally in a variety of applications like firefighting foams, surfactants, industrial emulsifiers, non-stick cooking utensils, paints, stain and water-resistant textiles and carpets, oil-repelling containers, etc. The C-F bond found in

PFAS compounds is known as the strongest bond in organic chemistry. This bond confers strong resistance to usual ecological transformation processes such as microbial, heat or chemical degradation (Meegoda et al., 2020; Nguyen et al., 2020; Wackett, 2021; Berhanu et al., 2023).

According to studies dating back nearly 2 decades, the probable route through which PFAS enter sewage sludge is mainly from industrial or residential wastewaters that flow into the wastewater treatment plants (Yu et al., 2009; Ma and Shih, 2010; Coggan et al., 2019; Semerád et al., 2020). PFAS in residential wastewaters results from the use of PFAS-containing products, such as personal care products, food packaging, carpets, and clothes (Schultes et al., 2018; Seltnerich, 2020; Zhu et al., 2021; Kumar et al., 2022; Rodgers et al., 2022). Additionally, extensive discharge of landfill leachate into municipal sewage systems has also been observed to increase PFAS concentrations in WWTPs (Masoner et al., 2020).

In a study conducted in the United States, PFAS concentrations in landfill leachate, influent and effluent samples collected from three landfill WWTPs were contrasted with similar samples from two WWTPs that received no landfill leachate (Thompson et al., 2022). The study concluded that the detection rate for all 73 PFAS



**FIGURE 2** Main options for the management of biosolids in selected countries. Source [China (2019) (Wei et al., 2020), United States (2019) (USEPA, 2021), Japan (2020) (Yosuke Matsumiya, 2020), Spain, Germany, United Kingdom, Poland, Netherlands, Türkiye, Romania, Austria and Sweden, (Eurostat, 2022), Canada (2012) (Canadian Council of Ministers of the Environment, 2012), South Korea (2022) (South Korea National Sewer Information System, 2022), Jordan (2011) (Batarseh et al., 2011), Iran, Brazil, Burkina Faso, Cameroon (2008) (UN-Habitat, 2008; The Australian and New Zealand Biosolids Partnership ANZBP, 2019) New Zealand, Australia and Colombia (2021) (Venegas et al., 2021), Chile and Israel (2017) (Organisation for Economic Co-operation and Development OECD, 2017), India, South Africa, Hong Kong (2017) (Sharma et al., 2017) Vietnam (2018) (ARCOWA, 2018) and UAE (2012) (Alshankiti et al., 2024)].

analyzed in leachate (92%) was higher than in influent (55%). The total PFAS content in leachate (93,100 ng L) was ten-fold higher than that of influent (6,950 ng L) and effluent (3,730 ng L). The leachate load contributions to PFAS in the WWTPs were found to vary from 0.78 to 31 g d<sup>-1</sup> (Thompson et al., 2022).

Investigations reveal that PFAS resists degradation in conventional WWTPs and are retained in both treated wastewater and biosolids; hence, the prevalence at different concentrations was observed (Table 1) from location to location (Meegoda et al., 2020; Kumar et al., 2022; Leung et al., 2022; Starnes et al., 2022; Brunn et al., 2023).

Exposure to PFAS has been associated with several health hazards, including decreased vaccination response in infants (Grandjean et al., 2017), heightened risk of type 2 diabetes in both men and women (Roth and Petriello, 2022), gestational diabetes in women (Birru et al., 2021), disrupted metabolic rate (Schillemans et al., 2021), reduced fertility (Rickard et al., 2022), reduced fetal development and obesity (Sevelsted et al., 2022; Liu et al., 2023), elevated risk of some cancers (Fenton et al., 2021; Ojo et al., 2021) and decreased immune system (Rappazzo et al., 2017).

This article systematically assessed the global generation of sewage sludge/biosolids, the diverse treatment methodologies employed, and the prevalence of contamination of per- and polyfluoroalkyl substances (PFAS) within these waste products globally. We also explored the factors influencing PFAS contamination in sewage sludge/biosolids and compiled the existing PFAS regulations or recommendations for land application.

## 2 Review methodology

To better understand the trends of global PFAS release into the environment through biosolid/sewage sludge applications, we reviewed multiple studies that addressed biosolid generation and management worldwide and the PFAS concentrations in biosolids from these regions.

A database search approach was implemented by independently selecting relevant keywords and terms and, in sequence, acquiring the appropriate publications from Web of Science, PubMed, ScienceDirect, Google Scholar, Scopus database, and federal and state records. The keywords were connected by the Boolean operators “AND” and “OR,” for instance, “PFAS in sewage sludge” OR “PFAS in biosolid AND “Countries” “Sewage Sludge AND Agriculture.” Other keywords include: “Biosolid generation in countries,” “PFAS variation in sewage sludge,” etc. Article exploration for this appraisal focused on articles published between 2003 and 2023. This extensive search was vital because it encompasses a substantial number of studies that will assist in validating the likely trends and movement of PFAS in the environment over time regarding PFAS use regulations. From the vast search results, the articles used in this review were selected based on their pertinence, thoroughness and focus on this review. [Supplementary Table S1](#), presents the list of abbreviations used in the review.

## 3 Survey of global biosolids generation and management—case studies

The annual global production of biosolids is experiencing a remarkable upswing, primarily due to the continuous expansion and technological progress of existing WWTPs. This upsurge can be directly linked to the escalating demand for water resources (Mohajerani et al., 2017). The global generation of biosolids is estimated at 100 to 125 million tons dry weight and is predicted to increase to 150–200 million tons dry weight by 2025 (Kumar et al., 2022). Countries are constantly looking for eco-friendly and sustainable ways to manage the increasing amounts of biosolids generated.

The global evaluation of biosolids generation and management is presented in [Figures 1, 2](#). The figures revealed that a significant percentage of biosolids end up in agricultural land applications or as landfills. Consequently, PFAS and other pollutants in these products are accessible for plant uptake and may enter our food chain.

Based on the United States Environmental Protection Agency reports in 2021, the United States generated 4.75 million dry metric tons of biosolids in 2019 (USEPA, 2021). Currently, the total biosolids generated in the USA are being managed in a variety of ways: land application (51%), landfill (22%), incineration (16%), additional management techniques (10%) and surface discharge (1%) (USEPA, 2021). Biosolids have been widely utilized in US agriculture since the early 1970s, supported by various regulations to promote their safe and beneficial application on land. Under stringent assessments, biosolids were no longer identified as wastes but as potential resources for nutrients, resulting in greater flexibility for biosolids management through agricultural application.

To evaluate the environmental risks linked to biosolids, the United States Environmental Protection Agency (USEPA), through a literature survey in 2019, assessed different contaminants present in biosolids generated across the United States (Zhen et al., 2017). The study found more than 500 potential pollutants in at least one instance, and variations of contaminants found in biosolids were observed to depend upon inputs to individual wastewater treatment facilities over time (USEPA, 2019).

A survey conducted between 2009 and 2019 on biosolid production from different provinces in China estimated that China generates 7.8 million tons of dry biosolids annually with a population of more than one billion (USEPA, 2019). Anaerobic digestion, thermal hydrolysis, composting and incineration are China's predominant sludge treatment technologies, with an estimated output of 6,944, 8,342, 11,250, and 27,122 t/dw in 2019, respectively. Zhen et al. (2017) affirmed that the status of sewage sludge management in China is significantly far from satisfactory, as more than 80% of sewage sludge generated is being discarded inappropriately. Under safety regulations for sanitary landfills in China, the threshold of sludge water content is 60% (Liu et al., 2021). Zhen et al. (2017) noted that even though landfill management is the most prevalent outcome for biosolids in China, most sewage sludge is disposed of directly after mechanical dewatering, with the sewage sludge still containing more than 80% moisture content and minimal compressive strength. Generally, 29.3% of the total biosolids generated in China are disposed of through land application, subsequently followed by incineration (26.7%) and sanitary landfills (20.1%), with much smaller quantities reused through their incorporation in building materials (15.9%) and various other methods (8.0%) (Wei et al., 2020; Chen et al., 2022).

Japan generates up to 2.0 million tons of biosolids annually, with reclaim and disposal rates climbing up to approximately 97%. Over 50 percent (52%) of Japan's biosolids are recycled into construction materials, while 31% is used in landfills, and 12% is digested anaerobically for bioenergy generation (Yosuke Matsumiya, 2020). Biosolid applications for agricultural purposes in Japan are relatively low, as farmers are concerned about the environmental risks of applying biosolids to agricultural land.

TABLE 1 Survey of PFAS concentrations in biosolids, influents and effluents from selected countries.

Continents	Location	Matrix	NO of WWTP	Period of sample collection	PFAS name	Concentration in biosolids (ng/g DW)	Conc. In influent (ng/L)	Conc. In effluent (ng/L)	References
North America	Canada	Biosolids	20	2009–2010	13 PFAAs	2.1–17,000	2.2–150	1.9–140	<a href="#">Guerra et al. (2014)</a>
	Canada	Biosolids	9	2015–2016	11 PFAAs	1,316 (expressed as F)	NA	NA	<a href="#">Lakshminarasimman et al. (2021)</a>
	Canada	Biosolids	20	2012–2017	22 PFAS	4.93–92.6	NA	NA	<a href="#">Letcher et al. (2020)</a>
	United States	Biosolids	94	2001	PFCAs	402	NA	NA	<a href="#">Venkatesan and Halden (2013)</a>
					PFSAs				
	United States	Biosolids	1	2005–2013	8 PFCA and 4 PFSA	22.5	NA	NA	<a href="#">Armstrong et al. (2016)</a>
	United States	Biosolids, Influent, Effluents	42	2018	24 PFAS compounds	195	50–800	50–4,800	<a href="#">Michigan Department of Environment et al. (2020)</a>
	United States	Biosolids, Influent, Effluents	6	2019	24 PFAS compounds	16–204	30–128	70–198	<a href="#">Tavasoli et al. (2021)</a>
	United States	Biosolids	8	2021	92 PFAS compounds	182–1,650	NA	NA	<a href="#">Thompson et al. (2022)</a>
	United States	Biosolids	38	2022	40 PFAS compounds	114–206	70–126	56–104	<a href="#">Schaefer et al. (2023)</a>
United States	Biosolids	190		24 PFAS	1–3,200	NA	NA	<a href="#">Link et al. (2024)</a>	
Europe	Germany	Biosolids	1,165	2008–2013	11 PFAAs	>500,000	NA	NA	<a href="#">Ulrich et al. (2016)</a>
	Germany	Sewage Sludge	NA	2010–2016	PFOA, PFOS	702 (PFOA) and 698 (PFOS)	NA	NA	<a href="#">Stahl et al. (2018)</a>
	Sweden	Sewage Sludge	4	2004–2017	79 PFAS compounds	50 and 1,124	NA	NA	<a href="#">Fredriksson et al. (2022)</a>
	France	Biosolids Composts	21	1976–2017	42 PFAS compounds	220	NA	NA	<a href="#">Munoz et al. (2022)</a>
	Spain	Biosolids	16	2011	8 PFAAs	0.21–120	NA	NA	<a href="#">Navarro et al. (2011)</a>
	Spain	Sewage Sludge	16	2010–2011	22 PFAS compounds	503–920	615–2,156	195–567	<a href="#">Campo et al. (2014)</a>
	Greece	Biosolids	2	2009–2010	18 PFAS compounds	237.2	717.5	201.1	<a href="#">Arvaniti et al. (2012)</a>
	Denmark	Sewage Sludge	10	2017	73 PFAS compounds	142	NA	45	<a href="#">Aro et al. (2021)</a>

(Continued on following page)

TABLE 1 (Continued) Survey of PFAS concentrations in biosolids, influents and effluents from selected countries.

Continents	Location	Matrix	NO of WWTP	Period of sample collection	PFAS name	Concentration in biosolids (ng/g DW)	Conc. In influent (ng/L)	Conc. In effluent (ng/L)	References
	Finland	Sewage Sludge	10	2017	73 PFAS compounds	129	NA	55.05	<a href="#">Aro et al. (2021)</a>
	Sweden	Sewage Sludge	10	2017	73 PFAS compounds	102	NA	163.41	<a href="#">Aro et al. (2021)</a>
	Sweden	Sewage Sludge	4	2004–2017	79 PFAS compounds	50 and 1,124	NA	NA	<a href="#">Fredriksson et al. (2022)</a>
	Norway	Sewage Sludge	10	2017	73 PFAS compounds	75	NA	44.04	<a href="#">Aro et al. (2021)</a>
	Italy	Sewage sludge	12	2018	PFOA	PFOA 2.5–3.5	NA	NA	<a href="#">Riva et al. (2021)</a>
					PFOS	PFOS 18.9–22.4			
Switzerland	Sewage Sludge	45	2008–2011	PFAAs	4–2,480	NA	191–1870	<a href="#">Alder and Van Der Voet (2015)</a>	
Asia	South Korea	Sewage Sludge	15	2010	15 PFAS compounds	0.8–1,400	610–2,100	NA	<a href="#">Kim et al. (2012)</a>
	South Korea	Biosolids, Influent, Effluents	25	2012	13 PFCAs	156–199	NA	269–561	<a href="#">Kwon et al. (2017)</a>
					PFSAs				
	China	Sewage Sludge	25	2011	C3–C14 PFAAs	126–809			<a href="#">Yan et al. (2012)</a>
	China		28	2010–2012	9 PFAS	1.12–413	NA	NA	<a href="#">Ruan et al. (2015)</a>
	Singapore	Sewage sludge	2	2006–2007	PFOA	PFOA 6.5–69.7	22–1,012.7	19.9–1,518.7	<a href="#">Yu et al. (2009)</a>
					PFOS	PFOS 30.7–702.2			
Thailand	Sewage sludge	2	2009	10 PFAAs	1,534.5	847.1	743.5	<a href="#">Kunacheva et al. (2011)</a>	
Hong Kong	Sewage sludge	3	2008	19 PFAS compounds	18.7–7,466.2	41.9–58.3	25.8–31.5	<a href="#">Ma and Shih (2010)</a>	
Africa	Nigeria	Sewage Sludge	10	2012	7 PFCAs and 3 PFSAs	0.01–0.597	NA	NA	<a href="#">Sindikou et al. (2013)</a>
	Kenya	Sewage Sludge	9	2013	10 PFAAs	0.098–0.683	0.9–9.8	NA	<a href="#">Chirikona et al. (2015)</a>
Australia	Australia	Biosolids	14	2014	9 PFAAs	5.2–150	0.98 to 440	21–560	<a href="#">Gallen et al. (2018)</a>
	Australia	Biosolids	19	2018	44 PFAS Compounds	4.2–910	NA	NA	<a href="#">Moodie et al. (2021)</a>

Based on a study by Collivignarelli et al. (2019), Europe generates more than 10 million tons of dry sewage sludge annually, with Germany, the United Kingdom, France, Spain, and Poland being the major contributors. Similar to biosolid management in China, land utilization, either through direct application or after composting, as well as incineration, are the primary sludge management process in European countries. About 50% of the total biosolids generated in Europe are spread on agricultural land; 28% are incinerated, while 18% are disposed of via landfills (Eurostat, 2022). For instance, in Ireland, 99% of sewage sludge generated in 2018 from urban areas was reused in agriculture (Eurostat, 2022).

The residual part is disposed of through landfill cover (e.g., Sweden), reuse in forestry (e.g., Slovakia, Latvia, and Ireland), pyrolysis, and storage (e.g., Greece, Italy, and Poland) (Eurostat, 2022). The compost approach is largely utilized in some European agricultural countries such as Luxembourg, Slovakia, Estonia, Hungary, Lithuania, etc. Between 40% and 60% of sewage sludge produced in these countries is managed through composting (Eurostat, 2022).

In Türkiye, Croatia, and Malta, about 42%–100% of the total sewage generated is disposed of in landfills. In contrast, incineration has been the predominant biosolid management practice in countries such as Belgium, Netherlands, and Germany due to the strict regulation on the agricultural utilization of biosolids as a result of possible contamination of the biosolids (Gianico et al., 2021).

Based on a survey conducted by the Canadian Council of Ministers of Environment (CCME) in 2012, Canada generates 660,000 tons of dry biosolids yearly with no data on the management of the biosolids. To encourage the advantageous use of biosolids, the CCME proposed dewatering, drying, and nutrient reclaiming from wastewater as acceptable management methods to enhance the quality of the biosolids generated (Canadian Council of Ministers of the Environment, 2012). In 2015, about 28.8% of biosolids generated were incinerated, while 15% were used in landfill (Tessier, 2017). According to Tessier, disposal of biosolids via incineration is declining, while land application is rising in Canada (Venegas et al., 2021). This trend is related to regulatory influences and voluntary biosolids quality enhancements but has been drastically slowed down by recent media attention on the presence of PFAS in biosolids.

Due to the paucity of information from these regions and the lack of appropriate and sufficient wastewater treatment facilities in India, South America, and Africa, estimating the annual biosolid production and management is challenging. Biosolids legislation, generation, stabilization, and application are underdeveloped in these continents and countries.

In Latin America, the primary strategy for managing biosolids/sewage sludge predominantly involves recycling them on agricultural land. This trend is exemplified by Colombia, where, in 2018, seven cities and four districts collectively generated 250,172 dry metric tons of biosolids (Venegas et al., 2021). Notably, 65% of this quantity was employed in agricultural areas. However, several WWTPs in this country dispose of sewage sludge into the environment without proper stabilization (Venegas et al., 2021).

In some areas of Chile, the percentage of biosolids recycled to land increased to 75% between 2009 and 2017. Between 2007 and

2017, over 285,836 tons of biosolids were dumped on 15,423 ha of farmland in the Brazilian state of Paraná (Bittencourt et al., 2014; Venegas et al., 2021).

Data from the India Central Pollution Control Board sets sewage generated in India at 62,000 million litres per day (MLD); however, only 23,277 MLD are being treated, which represents 37% of total sludge generated (PFAS, 2019). In 2021, the percentage of treated sewage increased to 50%, with more treatment plants being constructed. However, the remaining 50% of untreated sewage sludge is indiscriminately discharged into the environment (CPCB, 2021).

For Africa, it is difficult to estimate the amount of sewage sludge generated because individual households are often for the most part responsible for managing their own sewage (especially in rural and sub-urban settlements). While individual households rely on on-site septic tank systems for safe treatment of their wastewater, industrial wastewater is primarily treated through advanced water treatment methods (Onu et al., 2023).

Based on the Australian and New Zealand Biosolids Partnership Report 2019, Australia generates approximately 371,000 dry tonnes of biosolids yearly. Application to agricultural land is the main disposal route (70%), while 19% is used for landscaping or land restoration, and the residual is used as landfill or discharged to the ocean (The Australian and New Zealand Biosolids Partnership ANZBP, 2019).

## 3.1 Variations in PFAS concentration across wastewater influent, effluent and sewage sludge/biosolids

### 3.1.1 PFAS variation in wastewater influent and effluent

Generally, high PFAS concentrations have been reported in wastewater influents globally, especially influent from industrial source (Lenka et al., 2021). Studies have revealed the possibility of observing higher PFAS concentrations in effluents compared to the corresponding influent in WWTPs, which could suggest the potential transformation of initially undetected precursor ions (Gallen et al., 2018). While some studies reported increased PFAS concentrations in effluents compared to influents (Gallen et al., 2018; Tavasoli et al., 2021), other studies reported divergent results with higher PFAS in influents compared to effluents (Arvaniti et al., 2012; Campo et al., 2014; Schaefer et al., 2023).

In a study conducted by Tavasoli et al. (2021), the concentration of 24 PFAS compounds in the influents and effluents from 6 WWTPs in the United States was monitored. The study recorded a higher PFAS concentration in effluents (70 and 198 ng L<sup>-1</sup>) compared to influents (30–128 ng L<sup>-1</sup>). The study also recorded a higher PFAS concentration in effluents compared to influents during warmer seasons. The seasonal difference (in terms of temperature) of the treatment basins was between 4°C and 11°C in March and 20°C–23°C in July, and the PFAS concentration in influents within these seasons was unchanged. The increased PFAS in effluent in warmer seasons was therefore ascribed to further PFAS biochemical transformations with increased basin temperature, which is in accordance with prior studies. The study suggested the presence of higher concentrations of



fluorotelomers and other PFAS precursors in the effluents that could be converted during biological treatment (Tavasoli et al., 2021).

Studies show that degradation of precursor PFAS in influents results in higher PFAA concentration in effluents. Houtz et al. (2018) reported 200 and 350 ng L<sup>-1</sup> for PFPeA and PFHxA, respectively, in wastewater influent from the US. Within 3 days of wastewater treatment, the concentration of these two PFAS in the effluent increased to 1,690 and 1,370 ng L<sup>-1</sup>, respectively. Sun et al. (2012) also reported an increase from 26,000 ng L<sup>-1</sup> (influent) to 28,000 ng L<sup>-1</sup> (effluent) for PFOA in wastewaters from China.

In a study conducted by Gallen et al. (2018), a targeted analysis of 9 PFAS was carried out on influents, effluents and biosolids samples collected in 2014 from 14 WWTPs in Australia. The 9 PFAS concentrations detected ranged from 0.98 to 440 ng/L, 21–560 ng/L and 5.2–150 ng/g in influents, effluents and biosolids, respectively (Gallen et al., 2018). The estimated annual levels of PFOA and PFOS in wastewaters are 65 kg and 26 kg, respectively, while the yearly loads for biosolids were 2 kg and 8 kg, respectively. The study reported about a 9.8–59-fold increase in the total PFAS concentrations in effluents (Gallen et al., 2018). The observed rise in PFAS concentrations in effluents supports the production of PFAS from larger precursor chemicals within WWTPs, consistent with conclusion from several research studies (Dinglasan et al., 2004; Xiao, 2017).

In a nationwide review of PFAS concentrations in United States wastewaters, PFOA upsurged by an average of 6.0 ± 1.6 ng/L from the influents to the effluents of WWTPs. At the same time, PFOS showed no appreciable change in concentration. This indicates that PFAS sorption to sludge is counterbalanced by the biotransformation of precursor chemicals (Thompson et al., 2022).

### 3.1.2 PFAS occurrence and concentrations in biosolids from different locations

The evaluation of PFAS concentrations in biosolids can be used as model variables to estimate human susceptibility to PFAS through agricultural products from lands receiving biosolids. Table 1 summarizes the detection and concentrations of PFAS in biosolids from different countries. Most of the articles reviewed in this study gave a historical trajectory of changes in PFAS concentrations over specific periods to determine the trends of PFAS in the environment.

While some studies affirmed no decrease in PFAS concentrations in sewage sludge over the study period despite regulations (Armstrong et al., 2016), others reported both a decline and an increase in concentrations (Ulrich et al., 2016; Fredriksson et al., 2022; Thompson et al., 2022). Alder and van der Voet reported that an average reduction of 77% for perfluorooctanesulfonic acid (PFOS) and 62% for perfluorooctanoic acid (PFOA) within 2008 and 2011 in sludge generated from Switzerland are possibly due to the implementation of restrictions on perfluoroalkyl acids PFAAs (Alder and Van Der Voet, 2015).

The levels of PFAS pollution in biosolids collected between 2004–2017 were investigated in Sweden (Fredriksson et al., 2022). The samples were collected from four WWTPs servicing between 2,500 and 656,000 population units in Sweden. The treatment facilities were linked to residents, industries, and hospitals. The study reported the detection of 79 PFAS with a total concentration of

50 and 1,124 ng/g dry weight. All biosolid samples in the study showed detectable concentrations of C4- and C8-FASA-based copolymers ranging from 1.4 to 22 ng/g d.w. The study also confirmed that across all tested biosolid samples, the precursors of PFCAs, such as PAPs, FTCAs, and FTSAs, were the predominant groups, accounting for 56% of the 79 anionic and neutral PFAS (Fredriksson et al., 2022).

Longer chain PFAS have been detected in biosolids with increased frequency and concentrations because of their enhanced sorption capacity compared to their detection in effluent waters (Gallen et al., 2018). Ulrich et al. (2016) investigated the trend of 11 PFAS concentrations in biosolids generated from 1,165 different WWTPs in Germany between 2008 and 2013. The study affirmed the prevalence of PFOS in 41% of all sludge samples with a concentration of up to 7,600 µg/kg dm followed by PFOA (1,043 µg/kg dm). Within 15 WWTPs, total PFAS concentration was estimated to be > 500 µg kg<sup>-1</sup> dm with biosolids from 71 treatment plants exceeding the permissible threshold of 125 µg kg<sup>-1</sup> dm set for ∑11 PFAAs by the Bavarian State Ministry of the Environment and Consumer Protection (Ulrich et al., 2016).

Kwon et al. (2017) investigated the release of PFAS from industrial wastewaters in South Korea. Influent, effluent, and sewage sludge samples were collected from 25 industrial WWTPs connected to paper, textile, metal, electronics, and chemical industries. The paper sector had the greatest median concentration of PFAS in influent samples (411 ng/L), followed by the chemical industry (228 ng/L), the electronics industry (91.4 ng/L), the metals industry (32.6 ng/L), and the textiles industry (15.9 ng/L). PFOA and PFOS were the dominant PFAS (49%–66%). Reduced PFAS concentrations were observed in the effluent and biosolids compared to the influent. However, PFOA persisted in both effluent and sewage sludge.

The flow of PFAS through Canadian sludges collected between 2015–2016 was estimated through a mass balance approach at an average ∑PFAS-F mass flow of 1,316 ng/g (expressed as fluorine atom) (Venkatesan and Halden, 2013). Letcher et al. (2020) studied the influence of side-chain perfluoroalkyl polymer degradation in Canadian sludges and its effect on the concentration of PFAAs in sewage sludge. The concentration of side-chain perfluorooctane sulfonamide-urethane polymer and side-chain perfluorobutane sulfonamide-urethane polymer were evaluated. The study revealed that the concentration of polymers with side chain C<sub>8</sub>F<sub>17</sub> varied from 1.1 to 105 ng/g d.w. while those with C<sub>4</sub>F<sub>9</sub> side-chains varied from 37.5 to 2051 ng/g d.w. However, the total concentrations of 22 other PFAS analyzed in the study ranged from 4.9 to 93 ng/g d.w., which is thirty times lower than the side chain perfluoroalkyl polymers (Letcher et al., 2020).

Data emanating from Hong Kong revealed PFOS concentrations of 7,305 ng/g d.w. in sewage sludge (Ma and Shih, 2010), while PFOA concentration was 190 ng/g in Korean sewage sludge (Kim et al., 2012).

Campo et al. (2014) evaluated the prevalence and distribution of 21 PFAS compounds in biosolid samples collected between 2010–2011 from 16 different Spanish WWTPs. The study reported concentrations of PFAS ranging from 503 to 920 ng/g d.w.

Over the past 2 decades, PFAS levels in biosolids generated from WWTPs across the United States have been assessed for PFAS

(Arvaniti et al., 2014). A countrywide study was conducted in the United States to assess 13 per- and polyfluoroalkyl substances (PFAS) in representative biosolids samples collected from 94 WWTPs out of the more than 16,000 WWTPs in the U.S (Venkatesan and Halden, 2013). The samples were stored by the US EPA since the 2001 National Sewage Sludge Survey. The study discovered that the most prevalent PFAS found in biosolids mixtures from 32 US states and the District of Columbia was perfluorooctane sulfonate (PFOS) followed by perfluorooctanoate (PFOA) and perfluorooctanoate (PFDA) with concentrations of  $403 \pm 127$  ng PFOS/g dry wt.,  $34 \pm 22$  ng PFOA/g d.w. and  $26 \pm 20$  ng PFDA/g d.w. The study assessed the amount of PFAS in US biosolids to be between 2,749 and 3,450 kg per year, of which 1,375 to 2,070 kg are applied to farmland, and 467–587 kg are disposed of in landfills. Since 2001, however, the types of PFAS detected in biosolids have evolved due to manufacturing changes that favour short-chain PFAS. Work by Armstrong et al. (2016) evaluated the trends of 12 PFAS in biosolid samples collected from WWTPs serving more than 2 million population in the Mid-Atlantic region of the US from 2005 to 2013. The maximum mean concentrations detected over the study 8 years period for perfluorononanoic acid (PFNA), perfluorooctanoic acid (PFOA), and perfluorooctanesulfonic acid (PFOS) were 25.1 ng PFNA/g dw, 23.5 ng PFOA/g dw, and 22.5 ng PFOS/g dw, respectively, with 2.5–5 times higher detection frequency compared to others. The study affirmed that the concentrations of PFAS remain stable in WWTPs despite the regulation on the use of these compounds.

In a new study, Schaefer et al. (2023) monitored both quantifiable and semi-quantifiable PFAS in influents, effluents and sewage sludge from 38 WWTPs across 23 states within the US. The study confirmed the detection of PFAS in all samples analyzed. PFAS were thus found in every stream at every facility. In the influent, effluent, and biosolids, the aggregate concentration of detectable PFAS concentrations quantified were  $98 \pm 28$  ng/L,  $80 \pm 24$  ng/L, and  $160,000 \pm 46,000$  ng/kg dw, which mainly consisted of PFAAs. The study also detected perfluorophosphonic acids (PFPAAs) in 100% of the biosolid samples (Schaefer et al., 2023).

Arvaniti and Stasinakis conducted a thorough survey to review 36 PFAS monitoring studies conducted from 2005 to 2015 on wastewater and biosolids generated from the United States, North Europe, Asia, the Mediterranean area, Canada and Australia. The review concluded that PFOS appears to be the most prevalent analyte from all the studies reviewed, with a concentration of up to 7,300 ng/g dw. In contrast, the maximum PFOA concentration recorded was 240 ng/g dw, whereas the concentrations of longer PFCAs (such as C9) varied up to 3,200 ng/g dw (Arvaniti and Stasinakis, 2015).

Targeted and non-targeted screening of PFAS in biosolids and composts collected between 1976 and 2017 from France revealed the dominance of PFOS, EtFOSAA, and cationic and zwitterionic electrochemical fluorination precursors to PFOS in biosolids from municipal biosolids and composts while zwitterionic fluorotelomers dominated the urban organic waste products (Munoz et al., 2022).

Through fluorine mass balance analysis, Aro et al. (2021) evaluated the PFAS concentrations in Nordic Countries, including WWTP effluents and sewage sludge from Denmark, Finland, Faroe Islands, Iceland, Norway, Sweden, and Greenland.

The study recorded the concentrations of  $\sum 73$  PFAS as follows: Greenland (effluent–116 ng/L, biosolid 11 ng/g), Iceland (effluent–59, biosolids 4.6 ng/g) Faroe Islands (effluent–40 ng/L, biosolids—NA), Norway (effluent–44 ng/L, biosolids–75 ng/g), Denmark (effluent–45 ng/L, biosolids–142 ng/g) Sweden (effluent–163 ng/L, biosolids –102 ng/g) and Finland (effluent–55 ng/L, biosolids–129 ng/g). This study confirmed the dominance of PFCA precursors, while diPAP accounted for 62% of  $\sum 73$  PFAS. The study further evaluated the concentration of extractable organofluorine (EOF) by combustion ion chromatography analysis in influent, effluent and sewage sludge. The  $\sum$ EOF recorded was relatively higher (324–1,460 ng F/L in effluent and 39–210 ng F/g dry weight in sewage sludge) than those recorded for the sum of individual PFAS. The study affirmed that 90% of the  $\sum$ EOF recorded could not be accounted for by the  $\sum 73$  PFAS concentration recorded in the study, indicating the presence of some oxidizable precursor PFAS in the wastewater stream (Aro et al., 2021).

In the United Kingdom, Rigby et al. (2021) evaluated the concentration of PFAS in biosolids recycled to agricultural soils in the UK. The analyzed biosolids were generated from both industrial and municipal WWTPs. The study reported concentrations ranging from 99 to 231  $\mu\text{g kg}^{-1}$  for  $\sum 9$  PFAS, with PFOS, PFDA and PFBS being the dominant species.

Despite the detection of PFAS, especially PFOA and PFOS across aquatic systems (Ololade et al., 2018; Ssebugere et al., 2020) and human samples (Hanssen et al., 2010) in Africa, the sources of these contaminants have not been precisely identified. Until now, little data on PFAS emanate from Africa, especially for PFAS concentrations in sewage sludge. Sindiku et al. (2013) have evaluated PFAS concentrations in sludge generated from 10 WWTP across the western part of Nigeria and affirmed that PFAS were detected in all industrial, domestic, and hospital sewage sludge analyzed. PFOS was confirmed to be the prevalent PFAS in all sludge samples, with concentrations varying from 101 to 540  $\mu\text{g kg}^{-1}$ .

The India PFAS Situation Report in 2019 stated that PFAS use in India is unregulated (PFAS, 2019) consequently causing a significant level of PFAS in rivers (Sharma et al., 2016), groundwater (Lapworth et al., 2018) and aquatic biota (Murakami et al., 2011). Indiscriminate disposal of sewage sludge can be linked to this PFAS trend in India, as less importance has been placed on PFAS detection in sewage sludge (Kumari et al., 2023).

Bangladesh, Lebanon, Egypt, Indonesia, Jordan, Malaysia, Nepal, Vietnam, Sri Lanka, and Thailand joined the Stockholm Convention regulations on PFOS in 2009. However, these countries only regulated PFOS, while other PFAS chemical species remain unregulated. Furthermore, the regulations on PFOS and PFOA have not been implemented in these nations. Consequently, reports from these countries revealed the prevalence of PFAS in human samples, air, land, water, and aquatic life (IPEN, 2019). To our knowledge, no data is available for PFAS concentration in biosolids generated from these countries.

Reports from South America revealed PFAS pollution of both drinking water, surface, and groundwater (Kaboré et al., 2018; Souza et al., 2022). In Brazil, the concentration of  $\sum 23$  PFAS in an urban area surface and groundwater was found to range from 11 to 17 ng  $\text{L}^{-1}$  and 22–718 ng  $\text{L}^{-1}$ , respectively (Stefano et al., 2023). However,

no data was found for assessing PFAS in the biosolids from these nations.

### 3.2 The prevalence of PFAAs and diPAPs in sewage sludge and biosolids

A number of studies reviewed have consistently reported increased levels of perfluoroalkyl acids (PFAAs) during the wastewater treatment plant (WWTP) processes. This phenomenon has been attributed to either the biotransformation or oxidation of polyfluoroalkyl precursor compounds within WWTPs (Eriksson et al., 2017; Coggan et al., 2019; Zhang et al., 2021; Thompson et al., 2022). As terminal degradation products of precursor PFAS compounds, PFAAs resist degradation in WWTPs owing to their stability to both chemical and biological degradation processes (Starnes et al., 2022). The bulk of these contaminants, particularly the short-chain PFAA, infiltrate the receiving waters through the treated wastewater (Ulrich et al., 2016). Hence, WWTPs are an important contributor of PFAAs into aquatic environments (Ulrich et al., 2016; Thompson et al., 2022).

The high concentrations of PFAS observed in biosolids have been ascribed to the partial adsorption of long-chain PFAAs to sewage sludge/biosolids, attributed to their elevated affinity for the proteins and organic components of the biowastes. This makes biosolids and sewage sludge a significant source of PFAAs and potential transfer to the terrestrial environment if applied to soils (Ulrich et al., 2016).

PFOS has been widely reported as the dominant PFAS in sewage sludge (Ulrich et al., 2016; Fredriksson et al., 2022; Thompson et al., 2022). Alder and van der Voet studied the predominance of PFOS in sewage sludge generated from 45 wastewater treatment facilities in Switzerland collected between 2008 and 2011. The study reported concentrations ranging from 4–2,480  $\mu\text{g kg}^{-1}$  for  $\Sigma\text{PFAAs (C}_4\text{–C}_8\text{)}$ , while PFOS was always the predominant PFAS detected with a concentration varying between 4 and 2,440  $\mu\text{g kg}^{-1}$  (Alder and Van Der Voet, 2015).

Semerád et al. (2020) evaluated 32 PFAS in sewage sludge sourced from the WWTPs of 43 communities in the Czech Republic from 2018 to 2019. Their analysis revealed overall PFAS concentrations ranging from 5.6 to 963 ng/g. Notably, PFOS emerged as the predominant PFAS with a concentration of 933 ng/g (Semerád et al., 2020). The study also detected GenX in 9 samples, validating the use pattern and prevalence of new PFAS in the environment.

In activated sewage sludge reactors, the high PFOS adsorption to sewage sludge has been explained by comparing the partition coefficients ( $\log K_d$ ) of PFOS and PFOA between sludge and wastewater. A slightly higher partition coefficient obtained for PFOS (3.4 L  $\text{kg}^{-1}$ ) compared to PFOA (2.6 L  $\text{kg}^{-1}$ ) validates higher PFOS in sewage sludge compared to PFOA (Sun et al., 2012). However, Arvaniti et al. (2012) obtained somewhat divergent  $\log K_d$  values (PFOS–2.8, PFOA–3.1), indicating that sludge properties are distinct and can affect the sorption of PFAS. To support this divergent report, an assessment of PFAS pollution in sewage sludge collected in 2011 from 25 WWTPs in Shanghai, China, revealed the presence of 14 perfluoroalkyl acids (PFAAs) with varying carbon chain length (C3–C14) at

concentrations ranging from 126 to 809 ng/g. Perfluorooctanoic acid (PFOA) was also affirmed to be the dominant PFAS in this study, with concentrations ranging from 23 to 298 ng/g (Yan et al., 2012).

Guo et al., 2010, recorded a PFAS concentration of 278–5,383 ng/g in sewage sludge collected from 16 WWTPs across China in 2004 and affirmed that the data represents a good distribution of PFAS in China (Guo et al., 2010). The study found varying concentrations of PFOS and PFOA across different locations, with some areas having higher levels of PFOA and others having higher levels of PFOS (Guo et al., 2010).

While older studies mainly focused on PFAAs with few exceptions, recent reports have pointed to the frequent detection and prevalence of an unexpected PFAS class called disubstituted polyfluoroalkyl phosphates esters (diPAPs). This group of PFAS is commonly used in personal care products, surface protection applications and as coating agents in food packaging. As far back as 2009, D'eon et al. (2009) detected diPAPs in paper fibers from paper mills collected from Ontario in 2002 and 2003. The study recorded diPAP concentrations ranging from  $34 \pm 30$  to  $2,200 \pm 400$  ng/g.

In a recent study, Thompson et al. (2023a) detected diPAPs in toilet papers collected from North America, South and Central America, Africa, and Western Europe. Out of the 34 PFAS analyzed by the study, only 3 PFAAs and 3 diPAPs were detected in the toilet paper samples, with diPAPs accounting for  $91\% \pm 8\%$  of the total mass of PFAS in the toilet paper samples. The detection of diPAPs in various products implies their potential occurrence in sewage sludge. Reports from Sweden (Eriksson et al., 2017), Hong Kong (Loi et al., 2013), Canada (D'eon et al., 2009), US (Thompson et al., 2023a), and Australia (Moodie et al., 2021) confirmed the occurrence and the prevalence of diPAPs in sewage sludge.

While D'eon et al. (2009) recorded a concentration ranging from  $47 \pm 22$  to  $200 \pm 130$  ng/g for diPAP in sewage sludge from Ontario, Canada. Thompson et al. (2023a) recorded concentrations ranging from 73 to 1,400 ng  $\text{g}^{-1}$  for  $\Sigma_3\text{diPAP}$  making up  $54\% \pm 15\%$  of the  $\Sigma_{92}\text{PFA}$  on average in sewage sludge sourced from Florida (US) in 2021.

Schaefer et al. (2022) investigated the occurrence of 54 PFAS compounds sourced from 5 WWTPs in the US. The study recorded a total PFAS concentration varying from  $323 \pm 14$  to  $1,100 \pm 44$  ng/g, with diPAPs being the predominant PFAS in the biosolids. Moodie et al. (2021) also confirmed the dominance of diPAPs in Australian sewage sludge with a mean concentration of 140 ng/g dw. Loi et al. (2013) reported a relatively high concentration of diPAPs in sewage sludge and influent samples from Hong Kong in 2012, contributing between 40% and 95% to the total PFAS concentration in these matrices. The prevalence of diPAPs at high concentrations in sewage sludge suggests their widespread utilization and consumption in daily life.

### 3.3 Drivers of PFAS variations in sewage sludge/biosolids

Globally, the manufacture and use of PFAS are governed differently from nation to nation. Variations in the utilization of

products containing PFAS and their precursors among nations ultimately produce dissimilarities in the occurrence of PFAS in their biosolids. Additionally, the configurations of the proximal wastewater treatment facilities and the sludge treatment methods can be a significant source of disparity. For instance, [Guerra et al. \(2014\)](#) confirmed that forming PFAAs in WWTP depends on both system temperature and treatment methods, with higher formation rates in biological systems operating with extended hydraulic retention times and increased temperature.

[Kim Lazcano et al. \(2019\)](#) studied the effect of different sewage sludge treatment processes. They affirmed that there was an increased PFAA concentration using heat treatment and composting sewage treatment methods, while the thermal hydrolysis process had no noticeable influence on PFAA concentrations ([Kim Lazcano et al., 2019](#)). The study concluded that sources of PFAS contribute more to PFAS concentrations in biosolids than treatment processes. [Thompson et al. \(2023b\)](#), in a recent study, confirmed the prevalence of PFAAs in biosolids after anaerobic digestion, heat treatment, composting and lime treatment of the sewage sludge. The study recorded a decrease from  $\Sigma_{92}$ PFAS = 1,650 ng/g in the raw biosolids to 584 ng/g in biosolids treated with anaerobic treatment and belt press dewatering (65% decrease). Anaerobic treatment with heat drying used in another WWTP increased the concentration of  $\Sigma_{92}$ PFAS in the treated biosolids by 102%. The study explained that the belt-press dewatering process mechanically eliminates (extracts) mobile PFAS alongside the water content, whereas heat drying eliminates water through evaporation. The heat treatment results in the absorption of dissolved PFAS in the solid fraction, increasing the concentration of PFAS due to PFAA precursor transformation ([Thompson et al., 2023b](#)). To support this claim, [Zhang et al. \(2021\)](#) reported an increased concentration of extractable PFAAs in hydrothermally treated sludge at 165°C for 0.5/2 h and 250°C for 0.5 h. A complete degradation of PFCAs and an increase in PFSAs were observed at 300°C. Adding  $\text{Ca}(\text{OH})_2$  to hydrothermal treatment eliminated PFAA precursors and all PFSAs at 300°C, but substantially increased extractable PFAAs, except PFHpA and PFHxS ([Zhang et al., 2021](#)).

In a more elaborate study, [Ebrahimi et al. \(2021\)](#) investigated the influence of several solution-specific characteristics (pH, ionic strength), sludge characteristics (organic matter, lipids and protein content), PFAS characteristics (chain length, functional group), treatment and sludge stabilization methods on PFAS partitioning to sewage sludge. The study recorded an elevated PFAS concentration in sewage sludge at three distinct pH values—6, 7, and 8. This trend of increased PFAS sorption in more acidic conditions was also reported in another study ([Zhang et al., 2013](#)). The high sorption of PFAS to biosolids at elevated pH has been attributed to the reduced negative surface charge of the sludge at more acidic pH levels ([Ebrahimi et al., 2021](#)).

At elevated cation concentrations, the study also showed an increased PFAS sorption to biosolids, with results supported by other studies ([Chen et al., 2012](#); [Zhang et al., 2013](#)). This was ascribed to bridging mechanisms in which a divalent cation uses electrostatic attraction to bind with the negatively charged surfaces of the biosolids and the functional group of an anionic PFAS ([Ebrahimi et al., 2021](#)). High PFAS sorption to biosolids has also

been linked to increased fluoroalkyl chain length, with PFSA compounds having greater sorption capacities compared to PFCA compounds of similar chain length ([Coggan et al., 2019](#); [Ebrahimi et al., 2021](#)).

### 3.4 Regulations and advisory levels of PFAS in sewage sludge/biosolids

The Stockholm Convention was adopted by the international community in 2004 as an environmental protection strategy against persistent organic pollutants (POPs), and PFOA was added to the list in 2009. The convention has been ratified by 152 countries on all inhabited continents.

Between 2016 and 2022, the EPA placed the upper limits for PFOA and PFOS in drinking water at 70 ng/L. Recently, new limits have been adopted at 4 ng/L each for both PFOA and PFOS, 10 ng/L for PFNA, PFHxS, and GenX and a summation risk index for PFNA, PFHxS, GenX, and PFBS, following their initial advisory levels showing that the scientific evidence that practically no PFAS exposure level is safe ([Sauvé et al., 2023](#); [USEPA, 2024](#)).

The upper limit of total PFAS in drinking water in Canada has been proposed to be 30 ng/L for  $\Sigma_{30}$  PFAS ([Sauvé et al., 2023](#)), while the European Union placed the upper limit at 100 ng/L for  $\Sigma_{20}$  PFAS and 500 ng/L for  $\Sigma_{\text{total}}$  PFAS in drinking water ([EU, 2022](#)), China established limits only for PFOS and PFOA at 40 and 80 ng/L, respectively, in drinking water ([Wang et al., 2023](#)). However, the upper limit for PFAS in biosolids suitable for application on agricultural land has hardly been addressed.

A literature survey revealed that many countries, including China, India, Singapore, Central and South America, Japan, South Korea, and Africa, have no existing regulations on the upper limit of PFAS in biosolids before disposal or land application. In the United States, the EPA has established Regional Screening Levels (RSLs) for six PFAS compounds in soils ([Table 2](#)). However, these RSLs serve as guidance levels and do not carry legal obligations. Aside from the federal regulations on PFAS in soil, several states in the US have issued state RSLs for residential and industrial soils ([Table 3](#)). As of the time of this report, only Maine, New York and Michigan State have an existing rule on a few PFAS compounds in biosolids to be applied on agricultural land and biosolid-amended soil ([Table 2](#)).

As an interim strategy in Michigan, The Environment, Great Lakes, and Energy (EGLE) has developed a protocol for WWTPs whose biosolids may be contaminated by PFAS through industrial discharges. Michigan State has stipulated that biosolids with  $\geq 50$  ng/g PFOS require remedial actions before reuse, while biosolids with  $\geq 125$  ng/g PFOS are prohibited from reuse on agricultural lands. A similar strategy is currently in place in New York State with a limit of 20 ng/g for PFOA and 20 ng/g for PFOS in biosolids requiring remediation before land application, while biosolids with a concentration of  $\geq 50$  ng/g for PFOA and  $\geq 50$  ng/g for PFOS should not be land applied ([Maine Department of Environmental Protection, 2024](#)).

In 2009, the upper limit for PFAS in biosolids and soil was set at 100 ng/g in Germany due to recorded PFAS pollution that was explicitly linked to the land application of biosolids.

TABLE 2 Upper Limits of PFAS in biosolids and soils.

Matrix	Country	Regulatory level ( $\mu\text{g}/\text{kg}$ )	Reference
Soils	Germany (2009)	$\Sigma\text{PFAS}$ –100	NICOLE (2016)
	USEPA	PFOS (130)	BCLP Client Intelligent (2023)
		PFOA (190)	
		HFPO-DA (230)	
		PFBS (19,000)	
		PFHXS (1,300)	
		PFNA (190)	
Canada (2021)	PFOS (10)	CCME (2021)	
Biosolids-amended soils	Australia	$\text{C}_9\text{-C}_{14}$ (10)	NEMP (2020)
		PFOS (4)	
		PFOA (1)	
	Denmark	$\Sigma_{12}\text{PFAS}$ (400)	NICOLE (2016)
	Netherlands	PFOS (0.9)	The Dutch National Institute for Public Health and the Environment RIVM (2019)
		PFOA (0.8)	
	Sweden	$\Sigma_{11}\text{PFAS}$ (3)	Swedishepa (2017)
Biosolids	Maine, US (2019)	PFBS (1,900)	Maine PFAS Report (2020); Miller (2022)
		PFOS (5.2)	
		PFOA (2.5)	
	Michigan, US	PFOS (125)	Michigan Department of Environment et al. (2022)

In 2019, the US's Maine Department of Environmental Protection established a PFAS Task Force to evaluate PFAS contamination throughout the state and make recommendations. The task force, in their final report, placed the upper limit of perfluorobutane sulfonic acid (PFBS), perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in biosolids at 1,900, 5.2, and 2.5  $\mu\text{g}/\text{kg}$  dry weight respectively (Maine PFAS Report, 2020). However, the agricultural land application of biosolids was later totally forbidden in Maine (Maine Bill, 1911).

Aside from the three regulations mentioned from Germany, Maine State, New York and Michigan State, all other global regulations emanating from different locations provide upper limits for a few PFAS in soils only, especially after biosolid application (Table 2). For instance, in Denmark (NICOLE, 2016), a limit of 400  $\mu\text{g}/\text{kg}$  has been established for PFAS in soils, while in Sweden, the limit is set at 3  $\mu\text{g}/\text{kg}$  for gardens and residential lands and 20  $\mu\text{g}/\text{kg}$  for commercial and industrial land use (Swedishepa, 2017). Netherlands chose a different strategy by setting a stricter limit on PFOS and PFOA in soils than other countries [0.9 and 0.8  $\mu\text{g}/\text{kg}$ , respectively (The Dutch National Institute for Public Health and the Environment RIVM, 2019)].

In Canada, the Environment and Climate Change released the Federal Environmental Quality Recommendations that proposed a 10  $\mu\text{g}/\text{kg}$  limit for PFOS in agricultural soils (CCME, 2021).

In Australia, the limits for  $\text{C}_9\text{-C}_{14}$  PFCAs, perfluoroalkyl sulfonamides, fluorotelomer sulfonic acids, PFOA and PFOS in

soil after biosolid application were set at 10, 1, 4, 4, and 1  $\mu\text{g}/\text{kg}$  by the Queensland End of Waste Code Biosolids in 2020 (End of Waste Code Biosolids, 2020).

## 4 Conclusion

There is a substantial correlation between a country's level of development, industrialization and the prevalence of PFAS pollution in the environment. Basically, the development level in each country and the eventual disposal of more than 50% of biosolids generated globally to land, either through landfill or application to agricultural land, appear to be the primary local contribution to PFAS in soils globally. Potential atmospheric inputs distributed over large areas still need to be better assessed as rain and dry deposition are certainly contributing to low level large scale distribution of PFAS over soils. Albeit it is difficult to determine the actual levels of PFAS in developing countries, given the paucity of relevant data in these areas. This review article revealed a global, ubiquitous presence of PFAS in biosolids and little to no limits on the presence of PFAS in biosolids aimed for land application and introduction into our agri-food production. This does present a potential threat from the release of PFAS into the environment through agricultural land application, landfill or indiscriminate disposal of biosolids that are not usually systematically monitored for their levels of PFAS. The scant data on the occurrence of PFAS in biosolids worldwide reflects a lack of

TABLE 3 State regulations for PFAS in soils across the United States.

State	PFAS substance	General soil level ( $\mu\text{g}/\text{kg}$ )	Residential soil level ( $\mu\text{g}/\text{kg}$ )	Industrial/commercial soil level ( $\mu\text{g}/\text{kg}$ )
Alaska	PFOA and PFOS	Arctic Zone: 2,200		
		Over 40-inch Zone: 1,600		
California	PFOA		28	370
	PFOS		110	1,500
Connecticut	PFOA + PFOS + PFNA + PFHxS + PFHpA	1,350		
Delaware	PFOS and PFOA	1,300		
Florida	PFOS and PFOA		1,300	25,000
Indiana	PFOS	3,000		
	PFOA and PFNA	5,000		
	PFHxS	30,000		
	PFBS	500,000		
Iowa	PFOS	0.48		
	PFNA	180		
	PFHxS	1,600		
	PFBS	18,000		
	PFOA	35,000		
	PFHxA	39,000		
	PFBA	61,000		
Maine	PFOS and PFOA	1,700		22,000
	PFBS	1,700,000		22,000,000
Michigan	PFOS	0.22		
	PFOA	350		
Minnesota	PFOS		41	560
	PFHxS		130	1,700
	PFOA		240	3,200
	PFBA		38,000	520,000
	PFBS		57,000	77,000
Nebraska	PFOA		320	1,500
	PFOS		3,200	150,000
Nevada	PFOA		2,350	70,100
	PFOS		1,560	46,700
	PFBS		19,000	701,000
New Hampshire	PFOS		100	600
	PFHxS and PFNA		100	900
	PFOA		200	1,300
New Jersey	PFNA		47	670
	PFOS		110	1,600

(Continued on following page)

TABLE 3 (Continued) State regulations for PFAS in soils across the United States.

State	PFAS substance	General soil level ( $\mu\text{g}/\text{kg}$ )	Residential soil level ( $\mu\text{g}/\text{kg}$ )	Industrial/commercial soil level ( $\mu\text{g}/\text{kg}$ )
	PFOA		130	1,800
	GenX		230	3,900
New Mexico	PFOS, PFOA, PFHxS		1,560	260,000
New York	PFOA		6.6	500
			600	
	PFOS		8.8	440
			440	
North Carolina	PFOS		25	330
	PFHxS		250	3,300
	PFOA		38	490
	PFNA		38	490
	HFPO-DA (GenX)		47	700
	PFBS		3,800	49,000
Pennsylvania	PFOA and PFOS		4,400	64,000
	PFBS		66,000	960,000
Texas	PFOA		600	490
	PFOS		1,500	1,500
Vermont	PFOS, PFOA, PFHpA, PFHxS, PFNA		1,220	14,360
Washington	PFNA	200		
	PFOA	240		
	PFOS	240		
	HFPO-DA (GenX)	240		
	PFHxS	780		
	PFBS	24,000		
Wisconsin	PFOA and PFOS		1,260	16,400

Source: BCLP, Client Intelligence (BCLP, client intelligent, 2023).

interest and recognition of the potential risks caused by PFAS in biosolids.

This review observed that only Germany, Maine, Michigan State, and the USA have established some form of limits for a few PFAS in biosolids intended for land application. Moreover, in countries with regulatory standards, there appears to be little uniformity in the approach taken to determine the limits for PFAS in biosolids or soils. For instance, while some countries adopt lower limits of  $\leq 10 \mu\text{g}/\text{kg}$  for PFOS or PFOA for soils or biosolid-amended soils (Canada, Netherlands, and Australia), others adopted a rather higher concentration  $\geq 100 \mu\text{g}/\text{kg}$  for the sum of FAS (Germany and the US).

A national database that records PFAS usage and its presence in consumer products, including foods, biosolids, and manufacturing and processes using PFAS can help track and prevent the release of PFAS into the environment. We must also

better regulate the use of PFAS and prevent their release into wastewater and biosolids and the potential transfer to our food chain and underground water.

We must also implement better monitoring programs to document the presence and concentration of PFAS in biosolids and their production, management, and end-of-life, considering PFAS and the other emerging contaminants they contain.

## Author contributions

TS: Conceptualization, Data curation, Formal Analysis, Investigation, Methodology, Validation, Visualization, Writing—original draft, Writing—review and editing. SS: Conceptualization, Project administration, Supervision, Validation, Visualization, Writing—review and editing.

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## Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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## Supplementary material

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fenvc.2024.1383185/full#supplementary-material>

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