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Sludge and the circular economy – the impact of PFAS

This briefing note describes the various pathways PFAS can take to accumulate in air, soil and plants. It identifies the main contributors of PFAS in waste water and how PFAS can end up in sewage sludge. We demonstrate how only a full PFAS ban can allow for the inclusion of sewage sludge in the circular economy through agricultural application and incineration while encompassing the nutrient recycling potential.

Per- and polyfluoroalkyl substances (PFAS) are contaminants of critical concern due to their persistence, their mobility and widespread distribution in the environment. Their bioaccumulation is linked to potentially harmful human-health impacts.

To reduce the problems associated with PFAS in sewage sludge in both the circular economy and in the urban water cycle it is necessary to restrict and phase out all PFAS.

The ongoing restriction process of PFAS in fire-fighting foams and the increasing voluntary phasing out of PFAS in consumer products are examples of this progress. Other initiatives to tackle PFAS at source are decoupling UWWTPs – from PFAS manufacturers, from industries using PFAS, from storm water, from airports and from the leachate water from waste sites – and the upcoming restriction of all non-essential PFAS.

Summary

Per- and polyfluoroalkyl substances (PFAS) are contaminants of critical concern due to their persistence, their mobility and widespread distribution in the environment. Their bioaccumulation is linked to potentially negative human-health impacts (see the Annex for more information on PFAS).

PFAS compounds may be found in urban waste waters and some of these PFAS have been shown to accumulate in sewage sludge. High concentrations of PFAS occur especially when WWTPs receive influent from PFAS manufacturers, PFAS-using industries, storm water from airports and fire-fighting sites or leachate from waste sites.

Reducing or eliminating PFAS at the source is the most efficient action to addressing concerns related to PFAS in treated waste water, reclaimed water, biosolids and other resources from the WWTPs.

Biosolids with high PFAS levels applied to agricultural land may lead to elevated PFAS levels in groundwater and uptake into the food chain.

PFAS have been found in sewage sludge from exclusively domestic waste water as well, in ranges from 73 to 97µg/kg in primary sludge, and from 176 to 3,390µg/kg in digested sludge (Higgins et al 2005).

We have set out the limit values for some PFAS for some European countries and the results of sludge analysis reporting in this table:

Country	Sludge analysis (µg/kg DM -Dry Matter-)		Limit value in legislation (µg/kg DM)		
PFAS group	PFAS 4[1]	PFAS 22[2]	PFOA+PFOS	PFAS 4	PFAS 22
Austria			100		
Denmark	11 (average)	18 (average)		10	400
Germany			100		
Norway		2.63-168 25.2 (median)	40		

Thus, land application of biosolids as a beneficial soil improver may potentially allow PFAS to enter surface water through runoff or to infiltrate groundwater.

Biosolids affected by heavy-loaded PFAS sources may not meet future requirements for agricultural and land application. If a general ban of PFAS is not implemented, even biosolids from low polluted PFAS sources complying with current and future requirements could, in the long term, lead to PFAS accumulation in the environment due to their persistence.

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[1] PFOA, PFOS, PFNA and PFHxS.
[2] See Section 8 on PFAS legislation.

Biosolids from waste water treatment are nutrient rich, and once treated and tested to meet the regulation standards, they often are applied to agricultural land as a fertiliser or soil improver. Analysis of the literature indicates a direct correlation between PFAS concentrations in soil and bioaccumulation in plants. Based on the observation of PFAA accumulation in crops for human consumption, it has been concluded that soils conventionally improved for nutrients with municipal biosolids not impacted by high concentrated PFAS waste waters are unlikely to be a significant source of long-chain PFAS exposure in humans (Blaine et al., 2014).

Knowledge gaps remain regarding the monitoring of PFAS in the sludge matrix, e.g. the lack of a standard analytical method, available everywhere and comparable for many PFAS[3] or for total PFAS, and low capacities in many places to monitor PFAS. Sampling and analysis in sewage sludge are less reliable and more expensive than in water. These aspects should be taken into consideration if PFAS is to be included in the revision of the Sewage Sludge Directive (expected in 2024).

As regards end-of-pipe solutions, there is no comprehensive, cost-effective PFAS removal treatment at the water phase and no technology that separates PFAS from sewage sludge apart from incineration. The incineration route presents uncertainties, since conventionally-used sewage sludge incineration at relatively low temperature does not destroy PFAS completely, but instead transforms PFAS compounds, which are partially volatilised and emitted to the atmosphere, while another share of PFAS remains in the ashes. The high strength of the C-F bond, which characterises PFAS, makes their treatment with other technologies less effective, since the fluorinated compounds are not completely degraded, but only the length of their chain is only reduced. Because of the thermal stability of this bond, even the incineration of fluorinated substances is very complex and does often not ensure complete thermal degradation. PFOS and PFOA are effectively destroyed at 800 to 1000°C and 2 seconds residence time, although PFOS and PFOA destruction might lead to the formation of shorter-chain PFAS. The complete destruction of PFAS only takes place through incineration at around 1400°C and appropriate residence time and mixing. Thus, important knowledge gaps remain regarding the fate of PFAS during incineration that need to be addressed.

To reduce the problems with PFAS in both the circular economy and in the urban water cycle - from drinking water to waste water and to lakes, rivers and seas - it is absolutely necessary to restrict and phase out all PFAS.

The ongoing restriction process of PFAS in fire-fighting foams and the increasing voluntary substitution and therefore phasing out of PFAS in consumer products are examples in this direction. Other initiatives to tackle PFAS at the source are decoupling UWWTPs - from PFAS manufacturers, from industries using PFAS, from storm water, from airports and from the leachate water from waste sites – and the upcoming restriction of all non-essential PFAS.

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[3] There are analytical methods available for around 40 PFAS.

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1. Introduction - Sources of PFAS in waste water

PFAS impart water and grease repellent properties to products. They are therefore used, among other things, for impregnating textiles, in food packaging, in slip coatings in frying pans and cookware, and in ski wax. PFAS are also used in cosmetics.

> PFAS are used in many everyday products as their water and grease repellant properties make them extremely versatile.

Although there is a lot of learning still to be done and research projects are ongoing to determine the sources, it is a fact that PFAS are usually found in urban waste waters. There is a background level of PFAS in many waste water treatment plants (WWTPs) from consumer and household products. Additionally, there are point sources of PFAS with high concentrations coming from PFAS manufacturers or downstream user sites, landfill leachate and airport and fire-fighting run-off.

Conventional treatment at WWTPs does not eliminate PFAS, although polyfluoroalkyl compounds, acting as precursors, can be degraded into shorter perfluoroalkyl compounds. Most of the PFAS in the influent remain in the WWTP effluent, while a share of them - around 25% was estimated in the Netherlands (STOWA, 2021-46, Dutch Foundation for Applied Water Research) - end up in the sewage sludge.

2. PFAS, pathways to and accumulation in soil and plants

2.1. Pathways to soil

The transport and fate of PFAS in the environment are being investigated at multiple scales, from that of individual contaminated sites to global surveys. To date, research has focused primarily on the occurrence and transport of PFAS in the atmosphere, surface water and groundwater.

Sha et al (2022) state that there are three major sources of PFAA in the atmosphere: (1) direct emission from manufacturing sources such as fluoropolymer plants (Armitage et al. 2006, 2009), (2) formation in the atmosphere via degradation from volatile precursors like fluorotelomer alcohols (FTOHs) (Young et al. 2010; Thackray et al. 2020), and (3) water-to-air transfer via sea spray aerosol (SSA) emission (Reth et al. 2011; Johansson et al. 2019).

Relative to the atmospheric pathway, Thackray et al. (2020) reported that the atmospheric source of PFCAs is 6–185 tonnes per year globally and 0.1–2.1 tonnes per year in the Arctic. This amount comes from direct emissions, but also from the degradation of fluorotelomers in the atmosphere. Farther from direct emission sources, fluorotelomer-based materials degradation becomes the most important source of PFCA that leads to the release of PFCAs (Russel et al. 2008), precipitation, and water irrigation (Karnjanapiboonwong et al. 2018).

PFAAs can travel a great distance in the atmosphere via sea spray aerosol (SSA) (Sha et al. 2022) before being deposited on the ground. For SSA particles, residence times in the atmosphere are thought to be in the range of 10 hours to 10 days, depending on the size of the PFAS substance, and they can travel distances estimated at 100 – 2000 km in this time (Lewis et al. 2004). The volatility of (some) PFAS is underestimated. The so-called 'sea spray' which can occur during aeration is something which is being analysed/studied in the PERFORCE3-project by KWR (NL).

A report from The Netherlands estimates that between 65 - 180kg of PFAS are discharged annually in surface waters via all WWTPs (STOWA 2021-46).

However, there are indications that soils also serve as a significant reservoir for PFAS. The potential importance of soil as a global reservoir for PFAS was first quantified by Strynar et al. (2012). Samples were collected from locations far from known PFAS-contamination sources including industries known to have used PFAS. Rankin et al. (2016) reported concentrations of 32 PFAS in surface soil samples collected from 62 locations across all continents. These results suggest that the atmospheric long-range transport (LRT) of neutral PFAS followed by oxidation and deposition are a significant source of PFCAs and PFAs in soils. Washington et al. (2019) used the Rankin et al. data to calculate global soil loadings for 8 PFAS. These results indicate that soil has the potential to be a primary reservoir for PFAS (Brusseau et al. 2020).

The results of transport modelling conducted at individual contaminated sites also indicate that soils and the vadose zone therefore can serve as a significant long-term secondary source of PFAS (Shin et al. 2011; Xiao et al. 2015; Weber et al. 2017). This can serve as a long-term contamination source to surface water, groundwater, the atmosphere, and biota (Brusseau et al. 2020).

2.2. Transport and fate of PFAS in soil

The fate of PFAS in the soil is a function of many parameters including soil pH, soil structure, clay content, organic matter content (OM), PFAS characteristics (long versus short chain), and climatic conditions (Cai et al. 2019).

Many PFAS, especially short-chain, are very mobile, belonging to the PMT (persistent, mobile and toxic) and vPvM (very persistent and very mobile) hazard classes. Their mobility allows them to pass from one environmental compartment to another (from air to soil, soil to water and water to air).

Research found that PFAS can negatively affect the soil functionality where it may disturb soil enzyme activity as well as change the microbial availability and damage the cellular structure.

The mobile PFAS compounds that are soluble in water, have the potential to leach into groundwater, particularly in areas with potential source and route pollution like PFAS manufacturers, landfills and waste treatment plants, if solutions to prevent pollution are not in place. PFAS occurrence and leaching were reported by many researchers around the world (Perkola et al. 2013; Yin et al. 2017; Gallen et al. 2018; Robey et al 2020). The potentially-leaching PFAS could be alarming in many cases where PFAS were detected at large depths (15m) below ground (Dauchy et al. 2019). Yet, the leaching speed and behaviour vary from one PFAS to another, which depends on the soil binding, retardation and adsorption capacity (Bolan et al. 2021). The leaching characteristics of PFAS compounds are a function of the chain length where short chains are more mobile than long ones (Ziyad et al. 2020).

Long chain PFAS tend to adsorb in the surface layer of soils, until the adsorption capacity of the soil is reached. Then they can migrate to deeper layers and contaminate groundwater bodies.

Additional research on PFAS adsorption and migration from soil to groundwater and how this can migrate into groundwater needs further investigation and modelling to account for the various PFAS concentrations in different groundwater conditions (Sthal et al. 2009). Soil contaminated with PFAS is a challenge to reverse since there is no definite remediation strategy to address them in situ.

Research found that PFAS can negatively affect soil functionality where it may disturb soil enzyme activity as well as change the microbial availability and damage their cellular structure (Cai et al. 2019; Cai et al. 2020). The same result was confirmed by Sun et al. (2016a) as soil contaminated with PFAS compounds had less bacterial diversity. OM seems to be the most significant controlling factor determining the PFAS toxicity level where the PFAS toxicity is inversely proportional to the soil OM content (Zhao et al. 2016; Li et al. 2017).

Soil stabilisation can be achieved using various reagents such as clay and Portland cement, although it does not eliminate PFAS, since it does not remove PFAS permanently (Sthal et al. 2009; Ziyad et al. 2020).

2.3. PFAS accumulation in plants

PFAS uptake by plants poses a direct human risk where the food chain represents a main risk pathway. Therefore, a toxicological risk assessment addressing the maximum allowed

levels threshold of 4.4ng/kg body weight as a tolerable weekly intake (TWI) referred to as the sum of PFOA, PFNA, PFHxS and PFOS, was identified by the European Food Safety Agency (EFSA).

While biosolids transfer nutrients such as nitrogen and phosphorus to fields, this practice also transfers PFAS to soils, water and crops (Yoo et al. 2009, 2011; Washington et al. 2010). The main concern about PFAS in soil is the potential PFAS release and carryover by plants as well as the possibility of PFAS leaching underneath soil layers and into groundwater. This carryover of PFOA and PFOS to plants was evident when the PFAS level in plants was proportionally related to the PFOA/PFOS in the soil (Lechner et al. 2011). PFAS uptake from contaminated soil by crops was reported (Stahl et al. 2009), where samples from rye grass, grain, and potatoes showed a high potential of PFAS transfer from soil to crops (Stahl et al. 2009).

Analysis of the literature indicates a direct correlation between PFAS concentrations in soil and bioaccumulation in plants. Furthermore, plant uptake largely changes with chain length, functional group, plant species and organ. Bioaccumulation of perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) have been found at low levels in peeled potatoes and cereal seeds, while mobile short-chain compounds can accumulate at high levels in leafy vegetables and fruits. Significant variations in PFAS build-up in plants according to soil improvers are also found, suggesting a particular interaction with soil organic matter (Ghisi et al. 2019). PFAS are absorbed by plants to different extents according to their concentrations, chain lengths, functional group, plant species and variety, growth media (hydroponics vs. soil), and soil and biosolid characteristics. In particular, the abundance and characteristics of soil organic matter are considered the most important factors. Once inside the plants, partitioning among organs depends on species and, particularly, on functional group and chain length. The C4 – C6 compounds, which have recently replaced C8 PFOA and PFOS in many industrial processes, appear to accumulate particularly in leaves and fruits, whereas the compounds with higher chain lengths tend to be more concentrated in roots.

Plant uptake and the distribution of PFAS in the edible parts of plants is influenced by many factors, such as individual contaminants, soil organic carbon content or differences between species (Navarro et al. 2017; Ghisi et al. 2019; Liu et al. 2019b).

However, owing to the variability of available data, the complexity of the interactions between PFAS and soil components and between PFAS and plants, further studies need to be undertaken in order to clarify the role of each factor influencing the uptake and distribution of these compounds within plants, so as to reduce human exposure to these potentially hazardous substances (Ghisi et al. 2019).

An important environmental concern is that the long-chain PFAS can bioaccumulate as they move through the food chain. Compounds with a perfluoroalkyl chain length ≥8 are generally more bioaccumulative than those with ≤7 (Martin et al. 2003; Conder et al. 2008). Studies indicated that crops grown on contaminated soils can accumulate PFASs, suggesting that this may also be a source of human exposure (Stahl et al. 2009).

The soil functions and interactions with plants and surface and ground water demand a close consideration regarding PFAS in order to preserve the health of the soils.

3. PFAS in sewage sludge

Various classes of perfluoroalkyl compounds (including PFCA), perfluoroalkane sulfonates (PFSA) and perfluoroalkane sulfonamide (FASA) have been found in sewage sludge. However, important knowledge gaps remain on the PFAS transformation and adsorption processes in the waste water treatment that lead to PFAS uptake in sewage sludge. Some of the major problems are the lack of a standard (ie: available everywhere and comparable) analytical method for many PFAS as well as the scarce capacities to monitor PFAS in many places. In addition, sampling and analysis in sludge matrix are less reliable and more expensive than in water.

Even the few existing studies on PFAS in sewage sludge struggle to compare results from one campaign to the other because detection limits keep improving and new analytical methods become available for additional PFAS, while many PFAS remain undetected.

In general, studies show that the most abundant PFAS in the liquid phase of waste water is also frequently detected in the sludge phase (Arvaniti et al. 2012; Campo et al. 2014; Guerra et al. 2014). During waste water treatment, several functional groups undergo biotransformation. There are many known precursors (e.g. perfluorooctane sulfonamidoethanol, FOSA, FOSAA, etc.), which could be microbially transformed to recalcitrant PFAS not only during waste water treatment but also in soil (Rhoads et al. 2008; Liu and Avendano, 2013; Avendano and Liu, 2015). Moreover, it was documented that FTOHs and FTSs could be transformed to short PFCAs and PFSAs by resident microbial species in activated sludge under aerobic as well as anaerobic conditions (Wang et al. 2011; Li et al. 2018). Treated sewage sludge may also carry high levels of PFAS (Venkatesan and Halden, 2013; Letcher et al. 2020). Anaerobic and aerobic digestion of mixes of secondary and primary sludge has been shown to increase the concentration of PFAAs, compared to untreated sludge (Yu et al. 2009; Guerra et al. 2014). Possible reasons include the degradation of precursor compounds, the decrease of volatile solids during digestion, and increased sorption capacity of the digested sludge (Guerra et al. 2014).

> Treated sewage sludge may also carry high levels of PFAS. Anaerobic and aerobic digestion of mixes of secondary and primary sludge has been shown to increase the concentration of PFAAs, compared to untreated sludge.

Partitioning of PFAS to sludge in the waste water treatment plant increases with increasing chain length (Coggan et al. 2019). A study in Germany reported that about one-tenth of the load of PFOA and about half of PFOS arriving with the influent to a WWTP ended up in sludge (Becker et al. 2008). The predominance of even-chain length PFCAs like PFOA, PFDA, and PFDoA has also been reported in digested sludge (Sun et al. 2011; Guerra et al. 2014). Several authors have noted a decreasing trend in sludge contamination by longer PFAS

(PFOS and PFOA especially) in Germany (2008 - 2013) and Switzerland (2008 and 2011) during recent years, which could be explained by the use of new short-chain replacements with similar properties (Alder and van der Voet, 2015; Ulrich et al. 2016). Research has demonstrated that short-chain PFAS can be equally environmentally persistent and are even more mobile in the environment and more difficult to remove from drinking water than long-chain PFAS (Cousins et al. 2016; Sun et al. 2016b; Zhang et al. 2019).

The differences in sludge contamination across countries could be caused by factors such as population density, lifestyle, different industry and PFAS legislation or diverse waste water technology (Semerad et al. 2020).

The major factor affecting sludge contamination of PFAS in Norwegian sludge is shown to be where the WWTPs receive untreated leachate water from landfills, storm water from airports and from firefighting sites. In addition to those sources, industrial waste waters from PFAS manufacturers and downstream user sites are reported as causing PFAS polluted biosolids in other places like Flanders and The Netherlands.

Other sources are of a more diffuse character, reflecting the occurrence of PFAS in domestic products used in households. Sewage sludge from these sources usually presents low levels of PFAS.

Sludge analysis in Denmark from 38 WWTPs shows average concentrations for the sum of 4 PFAS (PFOA, PFOS, PFNA and PFHxS) of 11µg/kg DM (dry matter) and 18µg/kg DM for the sum of 22 PFAS (see the list of PFAS substances in the PFAS legislation section). A Norwegian survey of 22 PFAS from 18 major WWTPs, where analyses were carried out on monthly combined samples, found from 2.63-168µg/kg in digested sludge, with a median of 25.2µg/kg. The highest values came from WWTPs with run-off from airports with firefighting training sites connected.

Studies in Sweden have shown that PFAS11 from households contribute about 0.016mg/person/day and that the load from households are about 70% of the total load in influent waste water to the municipal WWTP. PFBA was the dominant PFAS in waste water from households, maybe this is due to more frequent use of e.g. 6:2 FTS, which forms the degradation product PFBA." (Ref. Cecilia Press, Fredrik Davidsson, Liza Wellsjö, Emelie Grubbström, Adam Ridelius (2020) Hushållspillvattenundersökning – Provtagningar i referensområden 2017/2018. Page 15,26,27,30 Appendix 2).

4. Sludge use in agriculture: the impact of PFAS

Based on the observation of PFAA accumulation in edible crops, (Blaine et al. 2014) it was concluded that soils conventionally improved for nutrients with municipal biosolids (not impacted by PFAS industries) are unlikely to be a significant source of long-chain PFAS exposure to humans. These municipal biosolids will normally comply with existing limit values on PFAS in sewage sludge for agriculture applications. Nevertheless, in the long term, these small amounts of PFAS will accumulate in the environment given their extreme

persistency. To address this problem there is no other solution than a broad phasing out of PFAS.

The application and the reuse of sludge in farmlands from waste water treatment plants impacted by PFAS manufacturers and downstream users, landfills, airports and fire-fighting sites is one of the main pathways that contribute to soil contamination.

On agricultural land, a further important contribution of perfluorinated compounds comes from the use of contaminated sewage sludge as a soil conditioner (Sepulvado et al. 2011; Zareitalabad et al. 2013). PFAS may be introduced to the environment through the land application of biosolids as a beneficial soil improver, potentially allowing PFAS to enter surface water through runoff or to infiltrate into groundwater (Lindstrom et al. 2011). The potential effects on groundwater or surface water depend on the amount and composition of PFAS present in biosolids, soil properties, infiltration rate, and land application practices. PFAS concentrations can be elevated in surface and groundwater in the vicinity of agricultural fields that received PFAS-contaminated biosolids over an extended period (Washington et al. 2010). The most abundant PFAS found in biosolids (PFOS and PFOA) are the same as those found in WWTP effluent, although biosolids may also contain other long-chain PFAS (Hamid and Li. 2016). In Flanders, Aquafin reports PFOS, PFOA, PFDA, PFHxA and PFPA as the predominant PFAS at WWTPs. Although the transformation of polyfluorinated substances to PFAAs in land-applied biosolids has been suggested (Sepulvado et al. 2011), other evidence suggests that some polyfluorinated substances remain in biosolid-improved soils for many years (Rich et al. 2015). The application of municipal biosolids as a soil improver can result in a transfer of PFAS to soil (Sepulvado et al. 2011). These PFAS can then be available for uptake by some plants and soil organisms. There are indications that PFAS can enter the food chain through the use of biosolid-improved soil (Lindstrom et al. 2011; Blaine et al. 2013; Blaine et al. 2014a; Navarro et al. 2017). It is noted, however, that PFAAs present at one municipal biosolids application site were not found in grain grown in the application plot (Gottschall et al. 2017).

Studies have shown that both industrially contaminated (Washington et al. 2010) and domestic municipal biosolids (Sepulvado et al. 2011) application as soil improvers can transfer PFAAs to the soil, with higher application resulting in increased PFAA concentration (Sepulvado et al. 2011). Following the transfer to the soil from biosolids, PFAAs may either accumulate on the surface of soil, leach into the subsurface and/or be taken up by plants and organisms (e.g. soil invertebrates). Preferential leaching of short chain PFCAs (<C8) were observed in biosolid-improved soil cores at depths of 1.2m or more (Sepulvado et al. 2011; Washington et al. 2010), indicating the potential for contamination of groundwater resulting from the application of typical municipal biosolids to agricultural fields (Sepulvado et al. 2011). PFOA (12ng/L) and PFOS (17ng/L) were also detected in tile drainage of agricultural plots improved with municipal biosolids (Gottschall et al. 2010).

Plant uptake of PFAAs and translocation into various above-ground parts have been observed from biosolid-improved, PFAA spiked-soils and water for various vegetables (e.g. carrot, lettuce, tomato, cucumbers, spinach, celery, snap pea) and staple foods (e.g. wheat, maize, potato) (Stahl et al. 2009; Lechner and Knapp 2011; Blaine et al. 2013; Wen et al.

2014; Bizkarguenaga et al. 2016; Navarro et al. 2017). The bioaccumulation of PFAAs from biosolid-improved soil depends on the concentration of PFAAs, physiochemical properties of the analyte, soil type, plant species and physiology (e.g. transpiration rate, lipid and water content) (Blaine et al. 2013; Wen et al. 2014; Navarro et al. 2017). In general, higher uptake and accumulation of PFCAs were reported compared to PFSAs in some crops (e.g. lettuce) (Blaine et al. 2014b; Bizkarguenaga et al. 2016).

The bioavailability and bioaccumulation of PFAAs to soil organisms from biosolids-improved soil represent a pathway for PFAS to enter the terrestrial food chain.

With the concern about PFAS contaminants in sewage sludge, some states in the U.S. are requiring PFAS testing of biosolids (Vermont Department of Environmental Conservation, 2019) or have suspended agricultural application (Maine Department of Environmental Protection, 2019), while others that had banned agricultural application are now considering a more risk-based approach.

Alternatives to the agricultural application of biosolids - landfilling or incineration of waste water sludge - also have significant environmental drawbacks (Stoiber et al. 2020).

A Swedish report (Svenskt Vatten, 2014) tested for PFOS and PFOA in soils and sugar beet crops fertilised with a combination of sewage sludge and mineral fertiliser, and determined that they were not present in levels above the detection limits of the analytical methods applied. These detection limits were (in 2014) as high as 0.010mg/kg DM. The study is being repeated in 2022 with improved analytical methods and much lower detection limits.

The Swedish report illustrates the difficulties that have led to the absence of a PFAS baseline or background levels in soils, and to assess when soil is contaminated. To address this knowledge gap, establishing long-term monitoring and surveillance management are key to maintaining a sustainably safe agricultural application of biosolids, allowing for an understanding of the accumulation of PFAS in soils and potential transfer to plants.

5. Thermal treatment of sludge containing PFAS

The preliminary findings suggest that sludge incineration – as is currently used - does not completely destroy PFAS, but instead transforms PFAS compounds, which are partially volatilised and emitted to the atmosphere, while part of the PFAS remains in the ashes. There are ongoing research projects to analyse the fate of PFAS in sludge during incineration.

The best-known and most widely used thermal degradation technology of PFAS on an industrial scale is incineration, which consists of an energy-intensive process, aimed at the complete mineralisation of the molecules. Conventional incineration of waste and biosolids uses temperatures well below the 1,400 °C needed to destroy PFAS. Other technologies, like supercritical water gasification, are under research.

The high strength of the C-F bond, which characterises PFAS, makes their treatment with other technologies less effective since the fluorinated compounds are not completely degraded, but only the length of their chain is reduced.

Because of the thermal stability of this bond, even the incineration of fluorinated substances is very complex and does often not ensure complete thermal degradation.

Under the ideal conditions of a uniformly mixed waste sample, with adequate stoichiometry, temperature and residence time of fuel and oxidant, most organic compounds can be destroyed to any desired degree and without the formation of by-products, with the exception of the ones set by thermodynamics.

For the combustion of fluorinated compounds, the products imposed by thermodynamics are CO_2, H_2O, HF (Tsang et al. 1998) and sulphates (Khan et al. 2020), in the case of PFAS with a sulphonic functional group.

However, ideal conditions are never perfectly and continuously achievable in an incinerator, and the biggest problem of PFAS incineration is in the nature and quantity of by-products formed by their incomplete combustion.

Incomplete mineralisation due to inadequate temperatures or insufficient residence times results in the production of stable by-products of various short-chain PFAS, such as CF4, C2F6 and C4F8 (Horst et al. 2020), which, unfortunately are powerful GHG.

A 2- to 10-fold decrease in the concentration of PFAAs (e.g., PFNA, PFOA, PFOS, PFDA) in sludge has been reported following incineration (Loganathan et al. 2007). Although this observation was made in a limited number of samples, (Arvaniti et al. 2012). In general, lower concentrations of PFAS in thermally dried sludge compared to dewatered sludge are reported (Hamid and Li. 2016).

The results of mono-incineration at 850°C of sludge highly loaded with PFOA (87µg/kg DM) and PFOS (250µg/kg DM) showed that no PFOS nor PFOA were found in the flue gas (less than 15ng/m³), the end products filter ash, absorbents and REA-gypsum (<10µg/kg DM) or the wash water also no PFOA and PFOS (<25ng/l) (North Rhine-Westphalia PFAS study, 2011).

Short-chain PFAS require higher temperatures to achieve thermal destruction compared to long-chain compounds (Watanabe et al. 2016), so their formation during heat treatment can further complicate the goal of achieving complete and safe mineralisation.

As the life cycle analysis (LCA) of PFAS continues to evolve, many recent studies on their heat treatment foresee an increasingly broad spectrum of PFAAs and associated fluorochemicals as potential by-products of their thermal destruction (Watanabe et al. 2016, Watanabe et al. 2018, Aleksandrov et al. 2019), clearly highlighting that, despite the high temperatures involved, even the incineration process, if not correctly managed, may not completely degrade the PFAS.

PFAS incineration or thermal treatment is de facto happening, either directly for PFAS-based materials such as firefighting foam (Hogue, 2020), or indirectly through the incineration of waste containing PFAS such as textiles or biosolids (Huber et al. 2009; Taylor et al. 2014; California Department of Toxic Substances Control. 2019; Khan et al. 2020; Solo-Gabriele et al. 2020).

A study in Japan investigated the fate of PFAS during thermal reactivation of granular activated carbon with absorbed PFOA, PFOS and 6-carbon perfluorohexanoic acid, reporting that after treatment at 700°C, a significant portion of the original compounds was converted to volatile species that escaped the final analysis (Watanabe et al. 2018).

Most studies are limited to highly controlled laboratory conditions and report on a specific number or subset of analytes. Several of these studies report that PFAS incineration under controlled conditions does not release the most-studied PFAS such as PFOA (Yamada et al. 2005; Taylor et al. 2014; Aleksandrov et al. 2019). There are also reports that specific PFAS, like PFOA, PFOS, and related compounds can be broken down with incineration (Krusic and Roe. 2004; Vecitis et al. 2009; Khan et al. 2020).

The specific profiles of fluorinated organic compounds released depend on incineration temperatures and operating conditions (García et al. 2007; Wang et al. 2013).

6. The circular economy and the immediate need for control at source measures

To be able to reach a sustainable circular economy it is necessary to act in line with the TFEU principles of Art.191.2 – the Precautionary Principle, principles that preventive action should be taken, that environmental damage should be rectified as much as possible at the source and that polluters should pay to reverse any environmental damage they cause.

These principles should be reflected in the legislative and non-legislative approaches, solutions and the good practices used on cross-cutting topics to achieve the common vision of the Zero Pollution objectives.

> Our sector's contribution to the circular economy depends greatly on the quality of the sludge used. We therefore see the need to decouple UWWTPs from industries using or producing PFAS, from storm water from airports and from leachate from waste sites.

As mentioned in chapter 4, based on the observation of PFAA accumulation in edible crops, Blaine et al. (2014) concluded that soils conventionally improved for nutrients with municipal biosolids (not impacted by PFAS industries) are unlikely to be a significant source of long-chain PFAS exposure to humans. Nevertheless, the persistence property of PFAS will lead to an environmental accumulation of PFAS in the long term, and the biosolids affected by heavy loaded PFAS sources may prevent the circular alternatives for biosolids.

There is no PFAS removal treatment from the water phase, no separation technology of PFAS from sewage sludge apart from incineration and the incineration route presents strong drawbacks as explained in chapter 5.

We therefore see the need to decouple UWWTPs from industries using or producing PFAS, from storm water from airports and leachate from waste sites. Moreover, in order to reduce the problems with PFAS in both the circular economy and in the urban water cycle - from

drinking water to waste water and to lakes, rivers and seas - it is absolutely necessary to restrict and phase out all PFAS. As an immediate action, policy must promote the voluntary substitution of PFAS in consumer products among e.g. sport and outdoor retailers and eco labelled household chemicals.

7. EU legislation

The Council of the European Union underlines the increasing health and environmental concerns posed by highly persistent chemicals. Per- and polyfluoroalkyl substances require special attention, considering the large number of cases of contamination of soil and water - including drinking water – in the EU and globally, the number of people affected with a full spectrum of illnesses and the related social and economic costs. That is why the European Commission proposes a comprehensive set of actions to address the use of and contamination with PFAS. Those aim to ensure, in particular, that the use of PFAS is phased out in the EU, unless it is proven essential.

7.1. UWWTD, IED, SSD[4], POP, CLP and PFAS restriction process

The Urban Waste Water Treatment Directive (UWWTD) and the Industrial Emission Directive (IED) must promote the Control at Source Principle and ban all PFAS releases to the environment, and discharges into sewers as well.

The Sewage Sludge Directive regulates the use of sewage sludge in agriculture also by setting limits on the presence of heavy metals, but not of organic contaminants.

The results of the evaluation of the Sewage Sludge Directive, to be launched soon, and the results of subsequent studies could provide the opportunity to introduce limits for organic contaminants such as PFAS in the event of the review of the Directive, including the possibility to have a limit for "total PFAS" and/or for a group of specific PFAS.

The EU works with the Member States and all Parties to the Basel Convention in the development and review of guidelines for sound management of persistent organic pollutants (POP) waste. The European Commission will work on the adaptation of the relevant guidelines following any future listing of PFAS substances as POPs.

As indicated in the new Conservation Effects Assessment Project (CEAP), the Commission will amend Annex IV of the POPs Regulation and is considering the introduction of a limit value for PFOA and the possible review of the value for PFOS.

The classification of PFAS substances under CLP (Classification, Labelling and Packaging) has immediate effects on the classification of waste containing these substances. Developments under GHS (Globally Harmonised System of Classification and Labelling of Chemicals) and CLP relevant for PFAS could result in the need to further align the waste legislation with CLP.

Five European countries have started the process of a PFAS group restriction to phasing out all PFAS in non-essential uses.

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[4] PFAS staff working document: https://ec.europa.eu/environment/pdf/chemicals/2020/10/SWD_PFAS.pdf.

7.2. Land and soil[5] contamination

Unlike for other environmental compartments, there is no dedicated European legislation on soil quality, although the process for a new Soil Health Law has started. For large industrial sites, there are provisions in the Industrial Emissions Directive (IED) that relate to soil /groundwater protection and remediation. More generally, the Environmental Liability Directive establishes a framework for preventing and remedying environmental damage.

Member States can set their own rules and standards, but only very few have defined limit values for PFAS in soil.

In Norway, a revised report made for the national Environmental Protection Agency (EPA) suggests normative values for contaminated soils, lowering the value for PFOS from 0.1 to 0.002mg/kg and establishing a new one 0.011 mg/kg for PFOA.

The EU expert group on soil protection could consider how soil contamination from PFAS can be addressed at EU level with a targeted and proportionate risk-based approach within a binding legal framework. They could also exchange knowledge and experiences on the safe, sustainable and circular re-use of excavated soil. This would allow for further building on the experience and knowledge of Member States in tackling PFAS in soils (e.g. ongoing work in Germany, Italy, Netherlands and Belgium). The Zero Pollution Action Plan, the new Soil Health Law and the revision of the thematic strategy for soil protection could also provide a framework to address the concerns raised by PFAS contamination of soils.

8. PFAS legislation in some Member States - examples

Denmark introduced limit values on PFAS in sewage sludge for agriculture use in October 2021. The Danish regulation focuses on two categories:
- Sum of 4 PFAS (PFOA, PFOS, PFNA and PFHxS) maximum: 10µg/kg dry matter.
- Sum of 22 PFAS (PFBS, PFPS, PFHxS, PFHpS, PFOS, PFNS, PFDS, PFUnS, PFDoS, PFTrS, PFOSA, 6:2 FTS, PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA) maximum: 400µg/kg dry matter.

In **Germany**, the Sewage Sludge Ordinance (Klärschlammverordnung) of October 2017 indirectly stipulates a threshold value for PFASs for the soil-related recovery of sewage sludge by referencing the Fertilisers Ordinance (Düngemittelverordnung) of May 2015. The latter generally sets a threshold value of 100 µg/kg total PFOS plus PFOA for both source materials of fertilisers as well as the fertilisers themselves; where the concentration exceeds 50µg/kg this must be indicated on the label.

In **Austria**, there is a limit for PFAS in sewage sludge used on agricultural soils – limit values of 100µg PFOS+PFOA /kg (0.1 ppm).

In **Sweden**, developed guideline values take into account the risks posed by a contaminated land area for people staying in the area (health risks), as well as the risks that the pollution means for the environment within the area (soil environment) and downstream the area

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[5] Idem.

(groundwater and surface water).

The preliminary guideline value for PFOS in land for sensitive land use is 3µg/kg DM and 20µg/kg DM for less sensitive land use (Statens geotekniska institute Publikation 21, 2015).

In **Norway**, new limit values have been proposed: sum PFOS+PFOA: 40µg/kg DM.

ANNEX

Per- and polyfluorinated alkyl substances (PFAS) belong to a group of highly persistent pollutants with more than 5000 identified substances (Liu et al. 2019a). The structure of PFAS typically consists of a fluorinated hydrocarbon forming the hydrophobic moiety of the molecule and a functional group, which represents the hydrophilic part. According to the functional group, PFAS can be divided into sulfonates (PFSA), carboxylates (PFCA), sulphonamides (FASA), sulfonamido acetic acids (FASAA), fluorotelomer acrylates and sulfonates (FTA and FTS), and others (Buck et al. 2011). The strong carbon-fluorine bond gives PFAS unique physicochemical properties desirable for applications, such as thermal, chemical stability and amphiphilic behaviour (Paul et al. 2009). Nevertheless, the strength and stability of the carbonefluorine bond also make the hydrophobic part of PFAS highly resistant to degradation or biodegradation in the environment (Parsons et al. 2008; Merino et al. 2016). Only some of the functional groups of PFAS may undergo partial biodegradation/biotransformation under appropriate environmental conditions, making the parent molecule a precursor for the formation of daughter products (mainly PFCA and PFSA) that do not decompose further and accumulate in the environment (Liu and Avendano, 2013; Semerad et al. 2020).

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EurEau is the voice of Europe's water sector. We represent drinking water and waste water operators from 30 countries in Europe, from both the private and the public sectors.

Our members are 35 national associations of water services. At EurEau, we bring national water professionals together to agree European water sector positions regarding the management of water quality, resource efficiency and access to water for Europe's citizens and businesses. The EurEau secretariat is based in Brussels.

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