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REVIEW ARTICLE

Role of wastewater treatment plant in environmental cycling of poly- and perfluoroalkyl substances

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Abstract *– The role of wastewater treatment plants (WWTP) in environmental cycling of poly- and perfluoroalkyl substances (PFASs) through aqueous effluent, sludge and air emission has been critically reviewed here. Understanding the role of WWTPs can provide better understanding of global cycling of persistent PFASs and assist in formulating relevant environmental policies. The review suggests that WWTP effluent is a major source of perfluoroalkyl acids (PFAAs) in surface water. Land application of biosolids (treated sludge) has shown preferential bioaccumulation of short chain (<C7) PFAAs in various plant compartments, leading to possible contamination of the food cycle. Elevated air concentration (1.5 to 15 times) of ∑PFASs have been reported at the aeration tanks on WWTP sites, compared to reference sites not contaminated with WWTP emission. The air emission of neutral PFASs has important implications considering the long-range transport and subsequent degradation of neutral compounds leading to the occurrence of recalcitrant PFAAs in pristine remote environments. Research gap exist in terms of fate of polyfluroalkyl compounds (neutral PFASs) during wastetwater treatment and in aquatic and terrestrial environment. Considering the wide range of commercially available PFASs, measuring only perfluorocarboxylic acid (PFCA) and perfluorosulfonic acid (PFSA) can lead to underestimation of the total PFAS load derived from WWTPs. Knowledge of the various pathways of PFAS from WWTPs to receiving environments, outlined in this study, can help in adopting best possible management practices to reduce the release of PFASs from WWTPs.*

Keywords *– PFAA, PFOA, PFOS, sludge, wastewater, pathway*

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1. Introduction

Poly- and perfluoroalkyl substances (PFASs) are a diverse group of synthetic fluorinated compounds. PFASs are aliphatic compounds, where all H atoms, except those in the functional groups, attached to all C (perfluoroalkyl) or in at least one C (polyfluoroalkyl) have been replaced by F atoms (Fig. 1). Due to the unique surface active properties and very high chemical and thermal stability imparted by the C-F bonds, (Buck et al. 2011), PFASs are widely used in numerous consumer products (e.g. textiles, paper, non-stick cookware, carpets, cleaning agents etc.) and industrial applications including metal plating, fire-fighting foams, electronics production, photography etc. (Kissa 2001; Arvaniti et al. 2014). There is a growing concern over the persistence, bioaccumulation potential and possible adverse effects in animal and humans of some PFASs, notably perfluoroalkyl acids (PFAAs). Two of the most commonly detected PFAA, pefluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA), have been detected in water (several ng/L range), human and animal blood and tissue, soil and sediment (concentration range of pg/g to ng/g) (Ahrens, 2011; Bartell et al. 2010; Bossi et al. 2008; Chen et al. 2012; Houde et al. 2011; Zareitalabad et al, 2013). Due to these findings, PFOS has been listed under Annex B of the Stockholm Convention Treaty on persistent organic pollutants (POPs) since 2009, prohibiting its production and use, except for a few exemptions; while perfluorooctanoic acid (PFOA) is currently under review by the POPs Review Committee (Convention 2008).

Direct emissions of PFASs during manufacturing and industrial processes, as well as use and disposal of consumer products containing PFASs (as additives or impurity) release these compounds into wastewater streams. During wastewater treatment, polyfluoroalkyl compounds (often called precursors) can degrade into perfluoroalkyl compounds (PFAAs) as shown in Fig. 1.

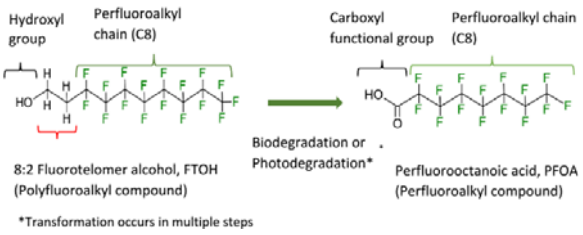

Fig. 1. Polyfluoroalkyl and perfluoroalkyl compounds (PFASs)

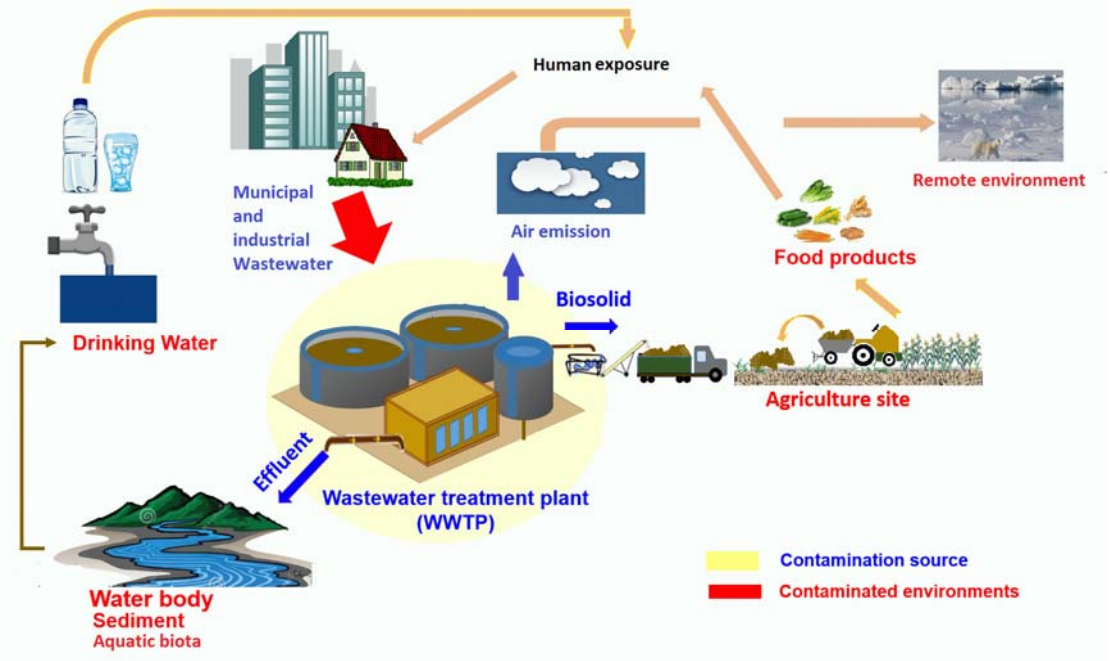

Fig. 2. Environmental pathways of poly- and perfluoroalkyl compounds (PFASs) from wastewater treatment plant (WWTP)

However, due to their recalcitrant nature, PFAAs are not efficiently removed during conventional wastewater and sludge treatment processes (Ahrens et al. 2011; Sun et al. 2011). PFASs are also frequently detected in wastewater treatment plant (WWTP) effluent at concentrations up to hundreds of ng/L (Arvaniti et al. 2014; Bossi et al. 2008; Higgins et al. 2005; Stasinakis et al. 2013) and are believed to be the major point source of these chemicals in the aquatic environment (Ahrens, 2011). The disposal of the solid or semi-solid product of WWTP (known as sludge) is also a cause of environmental concern. The environmental cycle and pathways of PFASs are illustrated in Fig. 2. A fraction of PFASs partition with sewage sludge during wastewater treatment, survive the subsequent sludge digestion processes and are detected in treated sludge (up-to 400 ng/g dw) (Campo et al. 2014; Guerra et al. 2014; Sun et al. 2011). The disposal of treated sludge (biosolids) on land acts as source of PFASs to enter surface water, groundwater (Zareitalabad et al. 2013) and the food chain (Lee et al. 2014; Yamashita et al., 2004). A limited number of studies also indicate that WWTPs act as a source of PFASs, notably semi-volatile and neutral PFASs, in the air (Ahrens et al. 2011; Weinberg, Dreyer, and Ebinghaus 2011; Shoeib et al. 2016), which can to be transported over great distances in the atmosphere and photodegrade, possibly leading to occurrence of PFAAs in remote and pristine environments (Yamazaki et al. 2016).

While WWTPs play a pivotal role in closing the cycle of water, the most valued natural resource, WWTPs also cycle contaminants (Pal et al. 2014). For the first time, this study critically reviews the environmental cycling of the PFASs through the aqueous effluent, sludge and air emissions from WWTPs to the receiving environment. Understanding the role of WWTPs can provide better understanding of global cycling of persistent PFASs (e.g., PFOS, PFOA) and assist in formulating relevant environmental policies. In addition to the well-studied perfluorinated compounds (e.g., PFAAs), the release of polyfluoroalkyl compounds are also considered here, as an increasing number of research papers prove them to be precursor compounds of PFAAs in the environment. Based on the review, existing knowledge gaps are identified, and future research directions are proposed in this study.

This paper is complementary to previous publications on PFASs that reviewed physiochemical properties (Ding and Peijnenburg 2013; Buck et al. 2011), microbial degradation (Liu and Avendano 2013), occurrence and fate in aquatic environment (Ahrens 2011), drinking water treatment processes (Rahman, Peldszus, and Anderson 2014) and WWTPs (Arvaniti and Stasinakis 2015).

2. PFASs in liquid effluent stream of WWTPs

2.1 Occurrence of PFAS in liquid stream of WWTP

PFAAs are routinely detected in wastewater from both municipal and industrial sources. Despite their phase-out by 3M, PFOA and PFOS are still two of the most frequently detected PFAAs in wastewater (Fig. 3), indicating their widespread past use and their continuing release (Guerra et al. 2014; Campo et al. 2014; Stasinakis et al. 2013; Arvaniti et al. 2012; Shivakoti et al. 2010). While the highest concentrations of PFOS and PFOA in municipal WWTP (in influent and effluent) were up to 465 ng/L and 638 ng/L, respectively (Arvaniti and Stasinakis 2015), much higher concentrations (>1000

ng/L) of PFOA and PFOS have been detected in industrial wastewater (Kim et al. 2012; Lin et al. 2014a). Additionally, municipal wastewater from highly urbanized and populated areas contains greater concentrations of PFAAs than rural areas (Sun et al. 2011). Other commonly-detected PFAAs in the liquid stream include perfluorohexanoic acid (PFHxA) (Guerra et al. 2014; Sun et al. 2012) perfluoropentanoic acid (PFPeA) (Arvaniti et al. 2012; Stasinakis et al. 2013) perfluorobutanoic acid (PFBA), (Campo et al. 2014), perfluoroheptanoic acid (PFHpA) and PFHxA (Shivakoti et al. 2010). Perfluorononaic acid (PFNA), perfluoroundecanoic acid (PFUnDA) and perfluorodecanoic acid (PFDA) have also been detected at lower frequencies and concentrations in some studies, indicating the prevalent production and use of shorter chain (≤ C8) PFCAs.

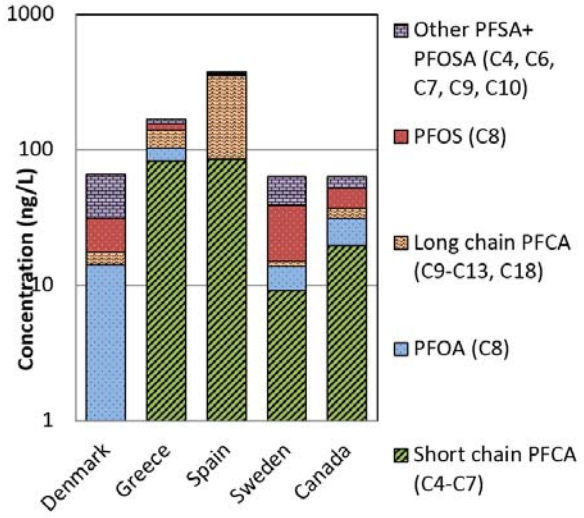

Fig. 3. Concentration of perfluoroalkyl compounds (PFAAs) in wastewater treatment plant effluent in various countries (Filipovic and Berger 2015; Guerra et al. 2014; Campo et al. 2014; Bossi et al. 2008)

2.2 Fate of PFASs in WWTP

Few studies have systematically investigated factors affecting the fate of these compounds along the conventional and/or advanced treatment train. The primary treatment aimed at physical settling of solids seems to provide very little or no removal of these compounds, possibly due to lower hydraulic retention time (HRT) and minimum biological activity of such units (Guerra et al. 2014; Shivakoti et al. 2010; Yu et al. 2009). Variable, poor and negative removal efficiency for most of the PFAAs were reported during biological treatment, as illustrated in Fig. 4 (Arvaniti et al. 2012; Campo et al. 2014; Guerra et al. 2014; Loganathan et al. 2007; Thompson et al. 2011; Yu et al. 2009). The observed increase in the secondary effluent concentration has been attributed to polyfluoroalkyl precursor degradation (fluorotelomer alcohols (FTOHs), perfluoroalkyl phosphates, or fluorotelomer sulfonates) in biological treatment processes. Biodegradation of polyfluoroalkyl compounds with microbial culture, activated sludge, soil and sediment has been reviewed by Liu et al. (2013). Despite an increase in removal efficiencies, no clear trend has been established so far. A recent study (Guerra et al. 2014) investigating the fate of 21 perfluoroalkyl acids across 20 Canadian WWTPs found that the effects of various treatment processes on formation of PFAAs differed statistically. In terms of high to low formation of PFAAs, the ranking was: advanced biological treatment with nutrient removal (median: 160%) > aerated/ facultative lagoon (150%) > secondary biological treatment (55%) > chemically assisted primary treatment (-1%) (Guerra et al. 2014). Greater formation of PFAAs (PFBA, PFHpA, PFNA and PFHxA) were observed with higher HRT and higher temperature in the summer, possibly due to increased associated microbial activities (Guerra et al. 2014). Another study (Chen et al. 2012) also reported relatively higher effluent concentrations of PFOS and PFOA in A2O treatment (Anaerobic / anoxic / oxic) technology compared to conventional activated sludge systems, biofilm process, and chemical flocculation (Chen et al. 2012). Although Yu et al. (2009) observed an increase in PFOA at higher solid retention times ((SRT)> 15 d), Guerra et al. (2014) did not find any correlation between formation of PFASs and SRT, mixed liquor volatile suspended solids, chemical oxygen demand or total suspended solids.

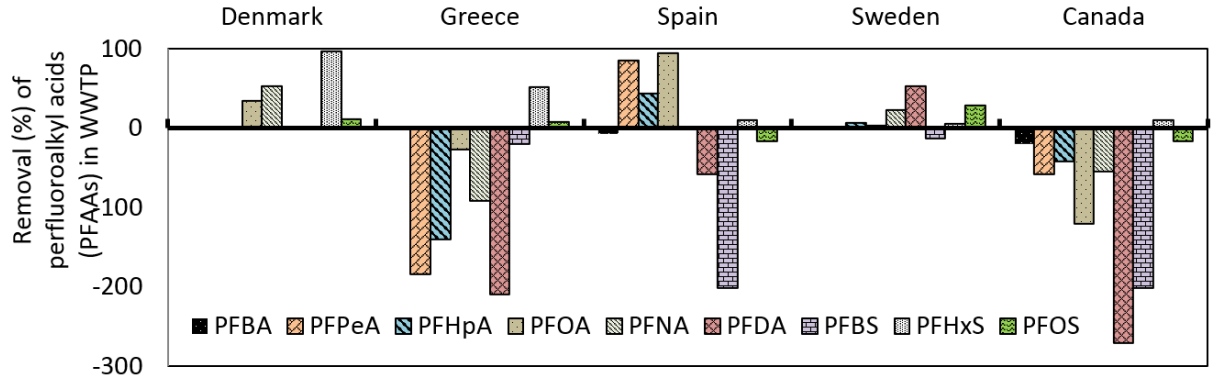

Fig. 4. Relative abundance of perfluoroalkyl compounds (PFAAs) in sewage sludge in various countries (Filipovic and Berger 2015; Guerra et al. 2014; Campo et al. 2014; Bossi et al. 2008)

Table 1: Occurrence of PFOA and PFOS (ng/L) in riverine and coastal water

	Concentration range in ng/L(median)			References
	PFOS	PFOA	Total PFAAs reported	
Surface river water				
Hanoi, Danang, Hue and Ho Chi Minh City, Vietnam	0.18–5.3 (0.27)[a]	0.09–18 (0.78)	1–17	(Duong et al. 2015)
Red River, Vietnam	0.21	0.16–0.52 (0.22)	0.2–1.2	(Duong et al. 2015)
Phong River, Thailand	<0.1–1.1	<0.2–8.8		(Lien et al. 2006)
Rivers in Japan	<0.1–191	<0.1–1.5		(Murakami et al. 2008)
Guangzhou, China	0.90–99	0.85–13		(So et al. 2007)
Yangzte River, China	<0.01–14	2.0–260		(So et al. 2007)
Pearl River Delta, China	0.02–12	0.24–16		(So et al. 2004)
Liao River, China	n.d.[b]–6.6 (n.d.)	n.d.–27.9 (11.5)		(Yang, Zhu, and Liu 2011)
Rhine, Germany	2.0–26	2.0–48		(Skutlarek, Exner, and Farber 2006)
Tennessee River, USA	16.8–114	<25–598		(Hansen et al. 2002)
European countries	(6)	(3)		(Loos et al. 2008)
Lake and stream water				
Taihu Lake, China	3.6–394 (5.8)	10.6–36.7 (19.5)		(Yang, Zhu, and Liu 2011)
Shihwa and Banweol, South Korea	2.24–651	0.9–62		(Rostkowski et al. 2006)
Lake Victoria , Africa	<.1–2.5	0.4–12		(Orata et al. 2009)
Coastal/bay water				
Moreton Bay, Australia	0.64–2.3	0.13–0.63	1.0–4.8	(Gallen et al. 2014)
East to South China Sea, China	<0.02–0.07	0.03–1.54		(Cai et al. 2012)
Sydney Harbour/ Parramatta River, Australia	7.5–21	4.2–6.4		(Thompson et al. 2011)
Estuarine and coastal of Korea	4.11–450 (28.5)	2.95–68.6 (14.7)		(Naile et al. 2010)
German Bight	0.69–3.95	2.92–7.83		(Ahrens, Felizeter, and Ebinghaus 2009)
South Korea	0.04–730	0.24–320		(So et al. 2004)
South China Sea	0.24–16	0.02–12		(So et al. 2004)
Hong Kong	0.73–5.5	0.09–3.1		(So et al. 2004)

[a]Median value reported in parenthesis; [b]not detected

2.3 PFAS pathways from WWTP effluent to environment

Studies have identified effluent from municipal and industrial WWTPs to be a major point source of PFASs in aquatic environments (Huset et al. 2008b; Ahrens et al. 2009; Möller et al. 2010; Sun et al. 2011; Lam et al. 2016). The concentrations in river water samples are approximately 5 - 12 times smaller than in WWTP effluents (Lam et al. 2016; Zareitalabad et al. 2013; Ahrens 2011). As shown in Table 1, PFOS and PFOA have been detected in tens to hundreds of ng/L concentration in river and lake water, with the higher concentration end representing streams receiving wastewater from fluorochemical industries (Wang et al. 2015; Zhao et al. 2015; Zareitalabad et al. 2013; Ahrens 2011; Möller et al. 2010; Huset et al. 2008a). The estimated mass flux of PFAAs (sum of C6-C9 PFCA and PFOS) from domestic wastewater source was 14.4 and 0.78 ton/y in Japan and Korea, respectively (Kim et al. 2012; Murakami et al. 2008). As the surface water is used as feed to the water supply system, PFAAs can then enter the urban water cycle (Pal et al. 2014). A recent study found that in a Swedish city (Bromma), the tap water was an important source of PFAAs to the WWTP influent, contributing >40% for each quantified perfluorosulfonic acid (PFSA) and up to 30% for PFCAs (Filipovic and Berger 2015), indicating the environmental recirculation of PFAAs in the urban water cycle. PFOA and PFOS have also been reported in sediments at concentrations up to 10 ng/g (Zareitalabad et al. 2013). However, very high concentrations (in the range of 50 - 70 ng/g) of PFAAs in sediments have been detected in the surroundings of fluorochemical plants (Lin et al. 2014a; Zhou et al. 2013).

Occurrence of PFAAs in freshwater biota (e.g., zooplankton, fish etc.) is well documented (Lam et al. 2016; Lam et al. 2014; Lin et al. 2014b; Houde et al. 2011). In general, the concentration of PFAAs has been observed to increase with increasing trophic level in the food chain in riverine ecosystems (Lam et al. 2016; Lam et al. 2014; Li et al. 2008). In addition to PFOS and PFOA, long-chain PFCAs, including PFNA, PFUnDA and perfluorotetradecanoic acid (PFTrDA), are often detected in biota tissues, owing to the more hydrophobic nature of long-chain PFAAs (Lam et al. 2014).

Despite the aforementioned studies on PFAAs, the fates of polyfluoroalkyl compounds and their degradation intermediates released with WWTP effluent are mostly unknown. A recent study (Ye et al. 2014) reported that total the PFCA (C4-C12) concentration increased by 16 ng/L in WWTP effluent following oxidation treatment, indicting the presence of PFCA precursors in the effluent. Under the same treatment conditions, the total PFCA concentration of the river water upstream and downstream

of the WWTP increased by 6.5 and 7.6 ng/L, suggesting a possible contribution of precursor compounds to the river water through the effluent discharge (Ye et al. 2014). This shows that the PFAS load from a WWTP would likely be underestimated if PFAA precursors are not measured.

3. PFASs in solid stream of WWTPs

3.1 Occurrence and fate of PFASs in sewage sludge

Various classes of perfluoroalkyl compounds, including PFCA, PFSA and perfluoroalkane sulfonamide (FOSA) have been detected in sewage sludge. In general, studies show that the most abundant PFASs in the liquid phase of wastewater are also frequently detected in the sludge phase (Arvaniti et al. 2012; Campo et al. 2014; Guerra et al. 2014). Additionally, long chain (>C8) PFCAs such as PFDA and perfluorododecanoic acid (PFDoA) (Sun et al. 2011), PFDA, PFUnDA and PFDoDA (Shivakoti et al. 2010), PFNA (Campo et al. 2014), and PFNA, PFDA (Guerra et al. 2014) are mainly detected in the sludge phase, as shown in Fig. 5. For PFOA and PFOS, two of the most widely studied PFAAs, concentrations up to 241 and 7304 ng/g dw have been reported (Kim et al. 2012).

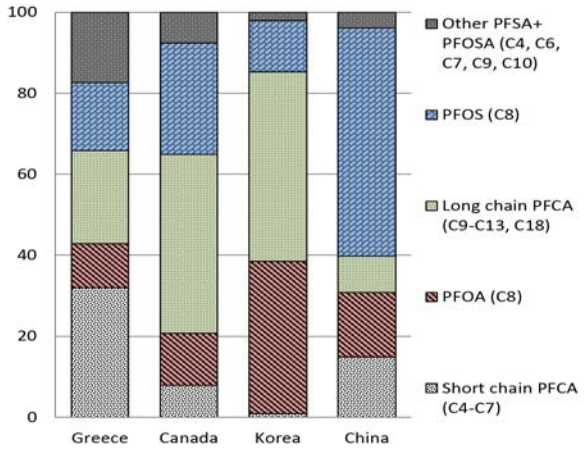

Fig. 5. Relative abundance of perfluoroalkyl compounds (PFAAs) in sewage sludge in various countries (Arvaniti et al. 2014b; Guerra et al. 2014; Kim et al. 2012; Sun et al. 2011a)

3.2 Fate of PFASs in sewage sludge treatment

Limited information is available regarding the fate of PFASs during sludge treatment processes (Arvaniti and Stasinakis 2015). Overall, anaerobic and aerobic digestion of mixed of secondary and primary sludge has been shown to increase the concentration of PFAAs, compared to untreated sludge (Guerra et al. 2014; Yu et al. 2009). Possible reasons include degradation of precursor compounds, decrease of volatile solids during digestion, and increased sorption capacity of the digested sludge (Guerra et al. 2014). Predominance of even-chain length PFCAs like PFOA, PFDA, and PFDoA has also been reported in digested sludge (Guerra et al. 2014; Sun et al. 2011). This is consistent with the biodegradation of FTOH leading to even-chain PFCAs under aerobic conditions (Higgins et al. 2005; Sinclair and Kannan, 2006). So far, only one peer-reviewed study investigated biodegradation of 6:2 and 8:2 FTOHs under anaerobic condition. Polyfluoroalkyl acids, e.g., fluorotelomer carboxylic saturated acids (FTCAs), fluorotelomer carboxylic unsaturated acids (FTUCAs) and x:3 acids were the major degradation products, as opposed to insignificant production of PFAAs (≤0.4% of 6:2 FTOH and 0.3% of 8:2 FTOH to PFHxA and PFOA, respectively) (Zhang et al. 2013).

In contrast to the above, a 2- to 10-fold decrease in 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) reported generally lower concentrations of PFASs in thermally dried sludge compared to dewatered sludge.

3.3 Cycling of PFASs through biosolids (treated sewage sludge)

A review by Clarke and Smith (2011) ranked PFAAs as the highest priority groups of emerging contaminants in biosolids that require additional research and monitoring. The criteria used by these authors in the assessment included environmental persistence, human toxicity, evidence of bioaccumulation in humans and the environment, evidence of eco-toxicity, and number and quality of studies focussed on the contaminant internationally (Clarke and Smith 2011). Previous studies have shown that both industrially contaminated (Washington et al. 2010) and typical municipal biosolids (Sepulvado et al. 2011) application as a soil amendment 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 biosolids-amended soil cores at depths of 1.2 m or more (Sepulvado et al. 2011; Washington et al. 2010), indicating the potential for contamination of groundwater resulting from application of typical municipal biosolids to agricultural fields (Sepulvado et al. 2011). PFOA (12 ng/L) and PFOS (17 ng/L) were also detected in tile drainage of agricultural plots amended with municipal biosolids (Gottschall et al. 2010).

Plant uptake of PFAAs and translocation into various above-ground parts have been observed from biosolids-amended, 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) (Bizkarguenaga et al. 2016; Wen et al. 2014; Blaine et al. 2013; Lechner and Knapp 2011; Stahl et al. 2009; Navarro et al. 2017). The bioaccumulation of PFAAs from biosolids-amended 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) (Wen et al. 2014; Blaine et al. 2013; Navarro et al. 2017). Both field and greenhouse studies on tomato and lettuce found

bioaccumulation factor (BAF=ratio of PFAA concentration in plant (ng/g dw) and in soil (ng/g dw)) greater than unity for C6 and shorter chain PFCAs, indicating accumulation in the plant tissues (Blaine et al. 2013). The BAFs for PFAAs in greenhouse lettuce decreased approximately 0.3 log units per CF_2 group (Blaine et al. 2013). Similarly, preferential bio-accumulation of PFAAs in the shoot have been reported for spinach (Navarro et al. 2017), wheat (Wen et al. 2014), celery, snap peas and radish (Blaine et al. 2014b). In general, higher uptake and accumulation of PFCAs were reported compared to PFSAs in some crops (e.g., lettuce) (Bizkarguenaga et al. 2016; Blaine et al. 2014a). Based on the observation of PFAA accumulation in edible crops, Blaine et al., (2014) concluded that soils conventionally amended for nutrients with municipal biosolids (not impacted by PFAA industries) are unlikely to be a significant source of long-chain PFAA exposure to humans.

The bioavailability and bioaccumulation of PFAAs to soil organisms from biosolids-amended soil represent a pathway for PFAAs to enter the terrestrial food chain. Using earthworms as model organism, PFAA bio-availability and bioaccumulation have been studied in biosolids-amended (Navarro et al. 2017; Wen et al. 2015; Rich et al. 2015), aqueous film-forming foam (AFFF)-contaminated (Rich et al. 2015) and PFAA-spiked soil (Zhao et al. 2013). BAF in the range of 1.5 – 4.99 (Navarro et al. 2017; Wen et al. 2015; Rich et al. 2015; Zhao et al. 2013) and 0.52 – 2 (Rich et al. 2015; Wen et al. 2015) have been reported for PFOS and PFOA, respectively. The BAF values increased with increasing carbon chain length for both PFCA and PFSAs (Navarro et al. 2017; Zhao et al. 2013). The accumulation of PFAAs is correlated positively with concentration of PFAAs and negatively with soil organic matter (Wen et al. 2015).

Following land application, polyfluoroalkyl compounds present in biosolids have been shown to undergo biodegradation and plant uptake. Lee et al. (2014) reported that, 6:2 polyfluoroalkyl phosphate diesters (diPAP) spiked with biosolids were biodegraded into corresponding fluorotelomer intermediates and C4-C7 PFCAs, following soil application. Accumulation of PFCAs present in the biosolids (0.1-138 ng/g wet weight (ww)) and those produced from 6:2 diPAP degradation (100-58000 ng/g ww) was also observed within 1.5 months of application (Lee, Tevlin, and Mabury 2014). Similarly, perfluorosulfonamide (PFOSA) spiked in compost degraded into PFOS after land application to carrot and lettuce plants (Bizkarguenaga et al. 2016). Both PFOS and PFOSA were reported to be taken up by these plants.

4. Air emission of PFASs from WWTPs

Occurrence of various classes of PFASs in WWTP ambient air (in pg/m^3 to ng/m^3 range) have been reported in a limited number of studies (Shoeib et al. 2016; Weinberg, Dreyer, and Ebinghaus 2011; Ahrens et al. 2011). As shown in Table 2, compared to reference sites, where air was presumably not contaminated by WWTP emission, measured PFASs concentrations were 1.5 to 15 times higher on WWTP sites. Generally, FTOHs were found to be dominant, accounting for 60 – 90% of total PFAS measured in activated sludge system (Shoeib et al. 2016; Weinberg, Dreyer, and Ebinghaus 2011; Ahrens et al. 2011). It is interesting to note that, depending on the type of treatment, the relative distribution of PFASs may change dramatically. For example, Shoeib et al. (2016) found that ∑PFAAs accounted for >70% of gas phase PFASs in a lagoon system treating wastewater. This distributional shift was likely caused by the longer hydraulic retention time (>3000 h compared to 5 – 16 h) of the lagoons, increasing degradation of the precursors (e.g., FTOHs) to PFAAs (Shoeib et al. 2016). PFASs emissions from WWTPs also correlated positively with wastewater inflow rate and population served, except for PFAAs, presumably indicating additional input due to precursor conversion (Shoeib et al. 2016). Using a Gaussian dispersion model, the total PFAS release to air was estimated to be 110 - 320 g/year/aeration tank in

Table 2: Concentration range of various classes of PFASs in air (pg/m^3) on WWTP sites

∑PFAAs[a]	∑FTOHs[b]	∑FOSAs, FOSEs[c]	Estimated input to air g/yr/tank	$\sum PFAS_{WWTP}/\sum PFAS_{reference\ site}$	Type of treatment	Reference
85 - 812	36 - 3766	<DL[d] - 24	37 – 110	1.5 to 6	Activated sludge	(Shoeib et al. 2016)
33 - 317	23 - 252	< DL - 18	3.5 – 13.4		Extended aeration	
8.5 - 398	26 - 93	<DL - 5	<2 – 6[f]		Facultative lagoons	
<DL – 10[e]	61 - 514	11 - 182	-	1.5 to 4	Activated sludge	(Weinberg, Dreyer, and Ebinghaus 2011)
214 - 408	1518 - 23706	21 - 124	320	3 to 15	Activated sludge	(Ahrens et al. 2011)

[a]perfluorocarboxylic and perfluorosulfonic acids; [b]fluorotelomer alcohols; [c]perfluoroalkane and N-alkyl perfluoroalkane sulfonamide and sulfonamidoethanols; [d]detection limit; [e]measured in particulates; [f]g/yr

activated sludge systems (Shoeib et al. 2016; Ahrens et al. 2011), an order of magnitude higher than the input (~ 12 g/year/tank) from extended aeration systems (Shoeib et al. 2016). The semi-volatile and neutral PFASs emitted from WWTPs can to be transported over great distances in the atmosphere and photodegrade (Ellis et al. 2004), possibly leading to occurrence of PFAAs in remote and pristine environments (Yamazaki et al. 2016).

5. Conclusions

The occurrence, fate and behavior of PFASs in wastewater treatment plants (WWTPs) has been reviewed here. While the occurrence of perfluoroalkyl (PFAA) compounds have been reported in many studies, precursor compounds (e.g., fluorotelomer alcohol, fluorotelomer sulfonates) and their intermediate degradation products have received much less attention so far. Despite the overall increase of PFAAs observed in treated wastewater following biological treatment, the factors affecting the PFAA formation process are not well understood and require further study. Similarly, research gaps exist in terms of the fate of PFASs during sludge treatment processes.

The environmental pathways of PFASs from WWTPs are also reviewed here. While, numerous studies have documented WWTPs to be a major source of PFASs in aquatic environments, fewer studies show that land application of treated sludge (biosolids) could also release PFASs to the environment. Historically PFOS and PFOA, and more recently other PFAAs, are found to be the focus environmental fate studies. Future monitoring studies should also include the known degradation intermediates of the precursor PFASs to avoid underestimation of the potential PFAA burden in an environmental compartment. The elevated ambient air concentration of primarily neutral PFSAs from WWTP sites were reported in a limited number of studies. While the estimated yearly air emission rate was at least 100 fold smaller (kg/y vs. tonne/y) compared to mass flux of PFASs in effluent, it nevertheless is important considering the long-range transport and subsequent degradation of precursor compounds leading to PFAA occurrence in pristine, remote environments.

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References

Ahrens, Lutz, Sebastian Felizetdoier, and Ralf Ebinghaus. 2009. "Spatial Distribution of Polyfluoroalkyl Compounds in Seawater of the German Bight." *Chemosphere* 76 (2): 179-184. http://dx.doi.org/10.1016/j.chemosphere.2009.03.052

Ahrens, Lutz, Merle Plassmann, Zhiyong Xie, and Ralf Ebinghaus. 2009. "Determination of Polyfluoroalkyl Compounds in Water and Suspended Particulate Matter in the River Elbe and North Sea, Germany." *Frontiers of Environmental Science & Engineering in China* 3 (2): 152-170. http://dx.doi.org/10.1007/s11783-009-0021-8

Ahrens, L. 2011. "Polyfluoroalkyl Compounds in the Aquatic Environment: A Review of their Occurrence and Fate." 13: 20-31. http://dx.doi.org/10.1039/C0EM00373E

Ahrens, Lutz, Mahiba Shoeib, Tom Harner, Sum Chi Lee, Rui Guo, and Eric J. Reiner. 2011. "Wastewater Treatment Plant and Landfills as Sources of Polyfluoroalkyl Compounds to the Atmosphere." 45: 8098-8105. http://dx.doi.org/10.1021/es1036173

Arvaniti, O. S., A. G. Asimakopoulos, M. E. Dasenaki, E. I. Ventouri, A. S. Stasinakis, and N. S. Thomaidis. 2014. "Simultaneous Determination of Eighteen Perfluorinated Compounds in Dissolved and Particulate Phases of Wastewater, and in Sewage Sludge by Liquid Chromatography-Tandem Mass Spectrometry." 6: 1341-1349. http://dx.doi.org/10.1039/C3AY42015A

Arvaniti, O. S., E. I. Ventouri, A. S. Stasinakis, and N. S. Thomaidis. 2012. "Occurrence of Different Classes of Perfluorinated Compounds in Greek Wastewater Treatment Plants and Determination of their Solid-Water Distribution Coefficients." 239: 24-31. http://dx.doi.org/10.1016/j.jhazmat.2012.02.015

Arvaniti, Olga S. and Athanasios S. Stasinakis. 2015. "Review on the Occurrence, Fate and Removal of Perfluorinated Compounds during Wastewater Treatment." 524: 81-92. http://dx.doi.org/10.1016/j.scitotenv.2015.04.023

Bizkarguenaga, E., I. Zabaleta, L. Mijangos, A. Iparraguirre, L. A. Fernandez, A. Prieto, and O. Zuloaga. 2016. "Uptake of Perfluorooctanoic Acid, Perfluorooctane Sulfonate and Perfluorooctane Sulfonamide by Carrot and Lettuce from Compost Amended Soil." *The Science of the Total Environment* 571: 444-451. http://dx.doi.org/10.1016/j.scitotenv.2016.07.010

Blaine, Andrea C., Courtney D. Rich, Erin M. Sedlacko, Katherine C. Hyland, Cecil Stushnoff, Eric RV Dickenson, and Christopher P. Higgins. 2014a. "Perfluoroalkyl Acid Uptake in Lettuce (Lactuca Sativa) and Strawberry (Fragaria Ananassa) Irrigated with Reclaimed Water." *Environmental Science & Technology* 48 (24): 14361-14368. http://dx.doi.org/10.1021/es504150h

Blaine, A. C., C. D. Rich, L. S. Hundal, C. Lau, M. A. Mills, K. M. Harris, and C. P. Higgins. 2013. "Uptake of Perfluoroalkyl Acids into Edible Crops Via Land Applied Biosolids: Field and Greenhouse Studies." *Environmental Science & Technology* 47 (24): 14062-14069. http://dx.doi.org/10.1021/es403094q

Blaine, Andrea C., Courtney D. Rich, Erin M. Sedlacko, Lakhwinder S. Hundal, Kuldip Kumar, Christopher Lau, Marc A. Mills, Kimberly M. Harris, and Christopher P. Higgins. 2014b. "Perfluoroalkyl Acid Distribution in various Plant Compartments of Edible Crops Grown in Biosolids-Amended Soils." *Environmental Science & Technology* 48 (14): 7858-7865. http://dx.doi.org/10.1021/es500016s

Bossi, R., J. Strand, O. Sortkjaer, and M. M. Larsen. 2008. "Perfluoroalkyl Compounds in Danish Wastewater Treatment Plants and Aquatic Environments." 34: 443-450. http://dx.doi.org/10.1016/j.envint.2007.10.002

Buck, Robert C., James Franklin, Urs Berger, Jason M. Conder, Ian T. Cousins, Pim de Voogt, Allan Astrup Jensen, Kurunthachalam Kannan, Scott A. Mabury, and van Leeuwen, Stefan P. J. 2011. "Perfluoroalkyl and Polyfluoroalkyl Substances in the Environment: Terminology, Classification, and Origins." 7: 513-541. http://dx.doi.org/10.1002/ieam.258

Cai, Minghong, Zhen Zhao, Haizhen Yang, Zhigao Yin, Qingquan Hong, Renate Sturm, Ralf Ebinghaus, Lutz Ahrens, Minggang Cai, and Jianfeng He. 2012. "Spatial Distribution of Per-and Polyfluoroalkyl Compounds in Coastal Waters from the East to South China Sea." *Environmental Pollution* 161: 162-169. http://dx.doi.org/10.1016/j.envpol.2011.09.045

Campo, J., A. Masia, Y. Pico, M. Farre, and D. Barcelo. 2014. "Distribution and Fate of Perfluoroalkyl Substances in Mediterranean Spanish Sewage Treatment Plants." 472: 912-922. http://dx.doi.org/10.1016/j.scitotenv.2013.11.056

Clarke, Bradley O. and Stephen R. Smith. 2011. "Review of 'emerging'organic Contaminants in Biosolids and Assessment of International Research Priorities for the Agricultural use of Biosolids." *Environment International* 37 (1): 226-247. http://dx.doi.org/10.1016/j.envint.2010.06.004

Convention, Stockholm. "What are POPs?", accessed September, 2016, http://chm.pops.int/TheConvention/ThePOPs/tabid/673/

Ding, Guanghui and Willie J. G. M. Peijnenburg. 2013. "Physicochemical Properties and Aquatic Toxicity of Poly- and Perfluorinated Compounds." 43: 598-678. http://dx.doi.org/10.1080/10643389.2011.627016

Ellis, David A., Jonathan W. Martin, Amila O. De Silva, Scott A. Mabury, Michael D. Hurley, Mads P. Sulbaek Andersen, and Timothy J. Wallington. 2004. "Degradation of Fluorotelomer Alcohols: A Likely Atmospheric Source of Perfluorinated Carboxylic Acids." 38: 3316-3321. http://dx.doi.org/10.1021/es049860w.

Filipovic, Marko and Urs Berger. 2015. "Are Perfluoroalkyl Acids in Waste Water Treatment Plant Effluents the Result of Primary Emissions from the Technosphere Or of Environmental Recirculation?" *Chemosphere* 129: 74-80. http://dx.doi.org/10.1016/j.chemosphere.2014.07.082

Gallen, Christie, Christine Baduel, Foon Yin Lai, Kristie Thompson, Jack Thompson, Michael Warne, and Jochen F. Mueller. 2014. "Spatio-Temporal Assessment of Perfluorinated Compounds in the Brisbane River System, Australia: Impact of a Major Flood Event." *Marine Pollution Bulletin* 85 (2): 597-605. http://dx.doi.org/10.1016/j.marpolbul.2014.02.014

Gottschall, N., E. Topp, M. Edwards, P. Russell, M. Payne, S. Kleywegt, W. Curnoe, and DR Lapen. 2010. "Polybrominated Diphenyl Ethers, Perfluorinated Alkylated Substances, and Metals in Tile Drainage and Groundwater Following Applications of Municipal Biosolids to Agricultural Fields." *Science of the Total Environment* 408 (4): 873-883. http://dx.doi.org/10.1016/j.scitotenv.2009.10.063

Guerra, P., M. Kim, L. Kinsman, T. Ng, M. Alaee, and S. A. Smyth. 2014. "Parameters Affecting the Formation of Perfluoroalkyl Acids during Wastewater Treatment." 272: 148-154. http://dx.doi.org/10.1016/j.jhazmat.2014.03.016

Hansen, K. Johnson, Ho Johnson, JS Eldridge, JL Butenhoff, and LA Dick. 2002. "Quantitative Characterization of Trace Levels of PFOS and PFOA in the Tennessee River." *Environmental Science & Technology* 36 (8): 1681-1685. http://dx.doi.org/10.1021/es010780r

Houde, Magali, Amila O. De Silva, Derek C. G. Muir, and Robert J. Letcher. 2011. "Monitoring of Perfluorinated Compounds in Aquatic Biota: An Updated Review PFCs in Aquatic Biota." 45: 7962-7973. http://dx.doi.org/10.1021/es104326w

Huset, Carin A., Aurea C. Chiaia, Douglas F. Barofsky, Niels Jonkers, Hans-Peter E. Kohler, Christoph Ort, Walter Giger, and Jennifer A. Field. 2008a. "Occurrence and Mass Flows of Fluorochemicals in the Glatt Valley Watershed, Switzerland." *Environmental Science & Technology* 42 (17): 6369-6377. http://dx.doi.org/10.1021/es703062f

Huset, Carin A., Aurea C. Chiaia, Douglas F. Barofsky, Niels Jonkers, Hans-Peter E. Kohler, Christoph Ort, Walter Giger, and Jennifer A. Field. 2008b. "Occurrence and Mass Flows of Fluorochemicals in the Glatt Valley Watershed, Switzerland." *Environmental Science & Technology* 42 (17): 6369-6377. http://dx.doi.org/10.1021/es703062f

Kim, Seung-Kyu, Jong-Kwon Im, Young-Min Kang, Se-Young Jung, Yeong Lim Kho, and Kyung-Duk Zoh. 2012. "Wastewater Treatment Plants (WWTPs)-Derived National Discharge Loads of Perfluorinated Compounds (PFCs)." *Journal of Hazardous Materials* 201: 82-91. http://dx.doi.org/10.1016/j.jhazmat.2011.11.036

Kissa, Erik. 2001. *Fluorinated Surfactants and Repellents*. Boca Raton: CRC Press.

Lam, Nguyen Hoang, Chon-Rae Cho, Kurunthachalam Kannan, and Hyeon-Seo Cho. 2016. "A Nationwide Survey of Perfluorinated Alkyl Substances in Waters, Sediment and Biota Collected from Aquatic Environment in Vietnam: Distributions and Bioconcentration Profiles." *Journal of Hazardous Materials* (In Press). http://dx.doi.org/10.1016/j.jhazmat.2016.04.010

Lam, Nguyen-Hoang, Chon-Rae Cho, Jung-Sick Lee, Ho-Young Soh, Byoung-Cheun Lee, Jae-An Lee, Norihisa Tatarozako, Kazuaki Sasaki, Norimitsu Saito, and Katsumi Iwabuchi. 2014. "Perfluorinated Alkyl Substances in Water, Sediment, Plankton and Fish from Korean Rivers and Lakes: A Nationwide Survey." *Science of the Total Environment* 491: 154-162. http://dx.doi.org/10.1016/j.scitotenv.2014.01.045

Lechner, Mareike and Holger Knapp. 2011. "Carryover of Perfluorooctanoic Acid (PFOA) and Perfluorooctane Sulfonate (PFOS) from Soil to Plant and Distribution to the Different Plant Compartments Studied in Cultures of Carrots (Daucus Carota Ssp. Sativus), Potatoes (Solanum Tuberosum), and Cucumbers (Cucumis Sativus)." *Journal of Agricultural and Food Chemistry* 59 (20): 11011-11018. http://dx.doi.org/10.1021/jf201355y

Lee, H., A. G. Tevlin, and S. A. Mabury. 2014. "Fate of Polyfluoroalkyl Phosphate Diesters and their Metabolites in Biosolids-Applied Soil: Biodegradation and Plant Uptake in Greenhouse and Field Experiments." 48: 340-349. http://dx.doi.org/10.1021/es403949z

Li, Xuemei, Leo Wai Yin Yeung, Muqi Xu, Sachi Taniyasu, Paul KS Lam, Nobuyoshi Yamashita, and Jiayin Dai. 2008. "Perfluorooctane Sulfonate (PFOS) and Other Fluorochemicals in Fish Blood Collected Near the Outfall of Wastewater Treatment Plant (WWTP) in Beijing." *Environmental Pollution* 156 (3): 1298-1303. http://dx.doi.org/10.1016/j.envpol.2008.03.008

Lien, Nguyen Pham Hong, Shigeo Fujii, Shuhei Tanaka, Munehiro Nozoe, Wanpen Wierojanagud, Ann Anton, and Gunilla Lindstorm. 2006. "Perfluorinated Substances in Tap Water of Japan and several Countries and their Relationship to Surface Water Contamination." *Environmental Engineering Research* 43: 611-618. http://dx.doi.org/10.11532/proes1992.43.611

Lin, Angela Yu-Chen, Sri Chandana Panchangam, Yu-Ting Tsai, and Tsung-Hsien Yu. 2014a. "Occurrence of Perfluorinated Compounds in the Aquatic Environment as found in Science Park Effluent, River Water, Rainwater, Sediments, and Biotissues." *Environmental Monitoring and Assessment* 186 (5): 3265-3275. http://dx.doi.org/10.1007/s10661-014-3617-9

Lin, A. Y. C., S. C. Panchangam, Y. T. Tsai, and T. H. Yu. 2014b. "Occurrence of Perfluorinated Compounds in the Aquatic Environment as found in Science Park Effluent, River Water, Rainwater, Sediments, and Biotissues." 186: 3265-3275. http://dx.doi.org/10.1007/s10661-014-3617-9

Liu, J. X. and S. M. Avendano. 2013. "Microbial Degradation of Polyfluoroalkyl Chemicals in the Environment: A Review." 61: 98-114. http://dx.doi.org/10.1016/j.envint.2013.08.022

Loos, Robert, Giovanni Locoro, Tania Huber, Jan Wollgast, Eugen H. Christoph, Alfred De Jager, Bernd Manfred Gawlik, Georg Hanke, Gunther Umlauf, and José-Manuel Zaldívar. 2008. "Analysis of Perfluorooctanoate (PFOA) and Other Perfluorinated Compounds (PFCs) in the River Po Watershed in N-Italy." *Chemosphere* 71 (2): 306-313. http://dx.doi.org/10.1016/j.chemosphere.2007.09.022

Möller, Axel, Lutz Ahrens, Renate Surm, Joke Westerveld, Frans van der Wielen, Ralf Ebinghaus, and Pim de Voogt. 2010. "Distribution and Sources of Polyfluoroalkyl Substances (PFAS) in the River Rhine Watershed." *Environmental Pollution* 158 (10): 3243-3250. http://dx.doi.org/10.1016/j.envpol.2010.07.019

Murakami, Michio, Eiji Imamura, Hiroyuki Shinohara, Kentaro Kiri, Yuki Muramatsu, Arata Harada, and Hideshige Takada. 2008. "Occurrence and Sources of Perfluorinated Surfactants in Rivers in Japan." *Environmental Science & Technology* 42 (17): 6566-6572. http://dx.doi.org/10.1021/es800353f

Naile, Jonathan E., Jong Seong Khim, Tieyu Wang, Chunli Chen, Wei Luo, Bong-Oh Kwon, Jinsoon Park, Chul-Hwan Koh, Paul D. Jones, and Yonglong Lu. 2010. "Perfluorinated Compounds in Water, Sediment, Soil and Biota from Estuarine and Coastal Areas of Korea." *Environmental Pollution* 158 (5): 1237-1244. http://dx.doi.org/10.1016/j.envpol.2010.01.023

Navarro, Irene, Adrián de la Torre, Paloma Sanz, Miguel Ángel Porcel, Javier Pro, Gregoria Carbonell, and de los Ángeles Martínez, María. 2017. "Uptake of Perfluoroalkyl Substances and Halogenated Flame Retardants by Crop Plants Grown in Biosolids-Amended Soils." *Environmental Research* 152: 199-206. http://dx.doi.org/10.1016/j.envres.2016.10.018

Orata, Francis, Natalia Quinete, Friedrich Werres, and Rolf-Dieter Wilken. 2009. "Determination of Perfluorooctanoic Acid and Perfluorooctane Sulfonate in Lake Victoria Gulf Water." *Bulletin of Environmental Contamination and Toxicology* 82 (2): 218-222. http://dx.doi.org/10.1007/s00128-008-9543-1

Pal, Amrita, Yiliang He, Martin Jekel, Martin Reinhard, and Karina Yew-Hoong Gin. 2014. "Emerging Contaminants of Public Health Significance as Water Quality Indicator Compounds in the Urban Water Cycle." *Environment International* 71: 46-62. http://dx.doi.org/10.1016/j.envint.2014.05.025

Rahman, Mohammad Feisal, Sigrid Peldszus, and William B. Anderson. 2014. "Behaviour and Fate of

Perfluoroalkyl and Polyfluoroalkyl Substances (PFASs) in Drinking Water Treatment: A Review." *Water Research* 50: 318-340. http://dx.doi.org/10.1016/j.watres.2013.10.045

Rich, Courtney D., Andrea C. Blaine, Lakhwinder Hundal, and Christopher P. Higgins. 2015. "Bioaccumulation of Perfluoroalkyl Acids by Earthworms (Eisenia fetida) Exposed to Contaminated Soils." *Environmental Science & Technology* 49 (2): 881-888. http://dx.doi.org/10.1021/es504152d

Rostkowski, Pawel, Nobuyoshi Yamashita, Iris Man Ka So, Sachi Taniyasu, Paul Kwan Sing Lam, Jerzy Falandysz, Kyu Tae Lee, Seung Kyu Kim, Jong Seong Khim, and Sook Hyeon Im. 2006. "Perfluorinated Compounds in Streams of the Shihwa Industrial Zone and Lake Shihwa, South Korea." *Environmental Toxicology and Chemistry* 25 (9): 2374-2380. http://dx.doi.org/10.1897/05-627R.1

Sepulvado, Jennifer G., Andrea C. Blaine, Lakhwinder S. Hundal, and Christopher P. Higgins. 2011. "Occurrence and Fate of Perfluorochemicals in Soil Following the Land Application of Municipal Biosolids." *Environmental Science & Technology* 45 (19): 8106-8112. http://dx.doi.org/10.1021/es103903d

Shivakoti, Binaya Raj, Shuhei Tanaka, Shigeo Fujii, Chinagarn Kunacheva, Suwanna Kitpati Boontanon, Chanatip Musirat, STMLD Seneviratne, and Hiroaki Tanaka. 2010. "Occurrences and Behavior of Perfluorinated Compounds (PFCs) in several Wastewater Treatment Plants (WWTPs) in Japan and Thailand." *Journal of Environmental Monitoring* 12 (6): 1255-1264. http://dx.doi.org/10.1039/b927287a

Shoeib, Mahiba, Jasmin Schuster, Cassandra Rauert, Ky Su, Shirley-Anne Smyth, and Tom Harner. 2016. "Emission of Poly and Perfluoroalkyl Substances, UV-Filters and Siloxanes to Air from Wastewater Treatment Plants." *Environmental Pollution* 218: 595-604. http://dx.doi.org/10.1016/j.envpol.2016.07.043

Skutlarek, Dirk, Martin Exner, and H. Farber. 2006. "Perfluorinated Surfactants in Surface and Drinking Waters." *Environmental Science and Pollution Research International* 13 (5): 299. http://dx.doi.org/10.1065/espr2006.07.326

So, MK, Y. Miyake, WY Yeung, YM Ho, S. Taniyasu, P. Rostkowski, N. Yamashita, BS Zhou, XJ Shi, and JX Wang. 2007. "Perfluorinated Compounds in the Pearl River and Yangtze River of China." *Chemosphere* 68 (11): 2085-2095. http://dx.doi.org/10.1016/j.chemosphere.2007.02.008

So, MK, S. Taniyasu, N. Yamashita, JP Giesy, J. Zheng, Z. Fang, SH Im, and Paul KS Lam. 2004. "Perfluorinated Compounds in Coastal Waters of Hong Kong, South China, and Korea." *Environmental Science & Technology* 38 (15): 4056-4063. http://dx.doi.org/10.1021/es049441z

Stahl, T., J. Heyn, H. Thiele, J. Hüther, K. Failing, S. Georgii, and H. Brunn. 2009. "Carryover of Perfluorooctanoic Acid (PFOA) and Perfluorooctane Sulfonate (PFOS) from Soil to Plants." *Archives of Environmental Contamination and Toxicology* 57 (2): 289-298. http://dx.doi.org/10.1007/s00244-008-9272-9

Stasinakis, Athanasios S., Nikolaos S. Thomaidis, Olga S. Arvaniti, Alexandros G. Asimakopoulos, Vasilios G. Samaras, Akinranti Ajibola, Daniel Mamais, and Themistokles D. Lekkas. 2013. "Contribution of Primary and Secondary Treatment on the Removal of Benzothiazoles, Benzotriazoles, Endocrine Disruptors, Pharmaceuticals and Perfluorinated Compounds in a Sewage Treatment Plant." 463: 1067-1075. http://dx.doi.org/10.1016/j.scitotenv.2013.06.087

Sun, Hongwen, Xianzhong Zhang, Lei Wang, Tao Zhang, Fasong Li, Na He, and Alfredo C. Alder. 2012. "Perfluoroalkyl Compounds in Municipal WWTPs in Tianjin, China—concentrations, Distribution and Mass Flow." *Environmental Science and Pollution Research* 19 (5): 1405-1415. http://dx.doi.org/10.1007/s11356-011-0727-6

Sun, Hongwen, Fasong Li, Tao Zhang, Xianzhong Zhang, Na He, Qi Song, Lijie Zhao, Lina Sun, and Tieheng Sun. 2011. "Perfluorinated Compounds in Surface Waters and WWTPs in Shenyang, China: Mass Flows and Source Analysis." *Water Research* 45 (15): 4483-4490. http://dx.doi.org/10.1016/j.watres.2011.05.036

Thompson, Jack, Anthony Roach, Geoff Eaglesham, Michael E. Bartkow, Katelyn Edge, and Jochen F. Mueller. 2011. "Perfluorinated Alkyl Acids in Water, Sediment and Wildlife from Sydney Harbour and Surroundings." *Marine Pollution Bulletin* 62 (12): 2869-2875. http://dx.doi.org/10.1016/j.marpolbul.2011.09.002

Wang, Tieyu, Pei Wang, Jing Meng, Shijie Liu, Yonglong Lu, Jong Seong Khim, and John P. Giesy. 2015. "A Review of Sources, Multimedia Distribution and Health Risks of Perfluoroalkyl Acids (PFAAs) in China." *Chemosphere* 129: 87-99. http://dx.doi.org/10.1016/j.chemosphere.2014.09.021

Washington, John W., Hoon Yoo, J. Jackson Ellington, Thomas M. Jenkins, and E. Laurence Libelo. 2010. "Concentrations, Distribution, and Persistence of Perfluoroalkylates in Sludge-Applied Soils Near Decatur, Alabama, USA." *Environmental Science & Technology* 44 (22): 8390-8396. http://dx.doi.org/10.1021/es1003846

Weinberg, Ingo, Annekatrin Dreyer, and Ralf Ebinghaus. 2011. "Landfills as Sources of Polyfluorinated Compounds, Polybrominated Diphenyl Ethers and Musk Fragrances to Ambient Air." *Atmospheric Environment* 45 (4): 935-941. http://dx.doi.org/10.1016/j.atmosenv.2010.11.011

Wen, Bei, Longfei Li, Hongna Zhang, Yibing Ma, Xiao-Quan Shan, and Shuzhen Zhang. 2014. "Field Study on the Uptake and Translocation of Perfluoroalkyl

Acids (PFAAs) by Wheat (Triticum aestivum L.) Grown in Biosolids-Amended Soils." *Environmental Pollution* 184: 547-554. http://dx.doi.org/10.1016/j.envpol.2013.09.040

Wen, Bei, Hongna Zhang, Longfei Li, Xiaoyu Hu, Yu Liu, Xiao-quan Shan, and Shuzhen Zhang. 2015. "Bioavailability of Perfluorooctane Sulfonate (PFOS) and Perfluorooctanoic Acid (PFOA) in Biosolids-Amended Soils to Earthworms (Eisenia fetida)." *Chemosphere* 118: 361-366. http://dx.doi.org/10.1016/j.chemosphere.2014.08.009

Yamazaki, Eriko, Jerzy Falandysz, Sachi Taniyasu, Ge Hui, Gabriela Jurkiewicz, Nobuyoshi Yamashita, Yong-Liang Yang, and Paul KS Lam. 2016. "Perfluorinated Carboxylic and Sulphonic Acids in Surface Water Media from the Regions of Tibetan Plateau: Indirect Evidence on Photochemical Degradation?" 51: 63-69. http://dx.doi.org/10.1080/10934529.2015.1079113

Yang, Liping, Lingyan Zhu, and Zhengtao Liu. 2011. "Occurrence and Partition of Perfluorinated Compounds in Water and Sediment from Liao River and Taihu Lake, China." 83: 806-814. http://dx.doi.org/10.1016/j.chemosphere.2011.02.075

Ye, Feng, Masahiro Tokumura, Md Saiful Islam, Yasuyuki Zushi, Jungkeun Oh, and Shigeki Masunaga. 2014. "Spatial Distribution and Importance of Potential Perfluoroalkyl Acid Precursors in Urban Rivers and Sewage Treatment Plant effluent–Case Study of Tama River, Japan." *Water Research* 67: 77-85. http://dx.doi.org/10.1016/j.watres.2014.09.014

Zareitalabad, P., J. Siemens, M. Hamer, and W. Amelung. 2013. "Perfluorooctanoic Acid (PFOA) and Perfluorooctanesulfonic Acid (PFOS) in Surface Waters, Sediments, Soils and Wastewater - A Review on Concentrations and Distribution Coefficients." *Chemosphere* 91 (6): 725-732. http://dx.doi.org/10.1016/j.chemosphere.2013.02.024

Zhao, Shuyan, Lingyan Zhu, Li Liu, Zhengtao Liu, and Yahui Zhang. 2013. "Bioaccumulation of Perfluoroalkyl Carboxylates (PFCAs) and Perfluoroalkane Sulfonates (PFSAs) by Earthworms (Eisenia Fetida) in Soil." *Environmental Pollution* 179: 45-52. http://dx.doi.org/10.1016/j.envpol.2013.04.002

Zhao, Zhen, Zhiyong Xie, Jianhui Tang, Renate Sturm, Yingjun Chen, Gan Zhang, and Ralf Ebinghaus. 2015. "Seasonal Variations and Spatial Distributions of Perfluoroalkyl Substances in the Rivers Elbe and Lower Weser and the North Sea." *Chemosphere* 129: 118-125. http://dx.doi.org/10.1016/j.chemosphere.2014.03.050

Zhou, Zhen, Yong Liang, Yali Shi, Lin Xu, and Yaqi Cai. 2013. "Occurrence and Transport of Perfluoroalkyl Acids (PFAAs), Including Short-Chain PFAAs in Tangxun Lake, China." *Environmental Science & Technology* 47 (16): 9249-9257. http://dx.doi.org/10.1021/es402120y