



Mass flow of per- and polyfluoroalkyl substances (PFAS) in a Swedish municipal wastewater network and wastewater treatment plant

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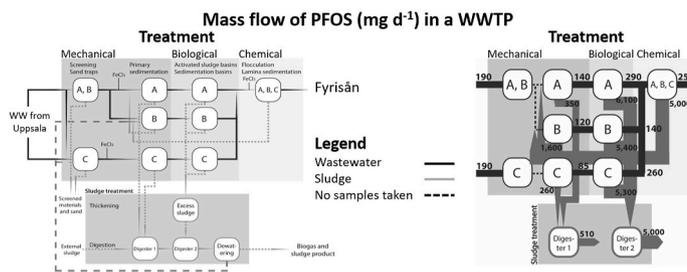
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HIGHLIGHTS

- PFAS data for a municipal wastewater network were used to identify sources.
- Short-chain PFAS dominated in wastewater, and long-chain PFAS in sludge.
- PFAS (de)sorption, recirculation, and transformation processes were assessed.
- Overall removal efficiency of PFAS in the WWTP was low.
- Discharge to the recipient was $43 \mu\text{g day}^{-1} \text{ person}^{-1}$ for $\sum_{26} \text{PFAS}$.

GRAPHICAL ABSTRACT



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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are ubiquitously distributed in wastewater, due to their numerous uses in industry and consumer products, but little is known of PFAS mass flows in municipal wastewater network systems and within wastewater treatment plants (WWTPs). This study assessed mass flows of 26 PFAS in a wastewater network and WWTP, to provide new insights into their sources, transport, and fate in different treatment steps. Wastewater and sludge samples were collected from pumping stations and the main WWTP in Uppsala, Sweden. PFAS composition profiles and mass flows were used to identify sources within the sewage network. Wastewater from one pumping station showed elevated concentrations of C₃–C₈ PFCA, likely caused by an industrial source, and two stations had elevated concentrations of 6:2 FTSA, probably originating from a nearby firefighter training facility. Within the WWTP, short-chain PFAS dominated in wastewater, whereas long-chain PFAS dominated in sludge. The ratio of perfluoroalkyl sulfonates (PFSA) and ethylperfluorooctanesulfonamidoacetic acid (EtFOSAA) to $\sum_{26} \text{PFAS}$ decreased during the WWTP process, likely due to sorption to sludge, but also transformation (EtFOSAA). Overall, PFAS were not efficiently removed in the WWTP, with mean removal efficiency of $10 \pm 68\%$ for individual PFAS, resulting in discharge of $7000 \text{ mg d}^{-1} \sum_{26} \text{PFAS}$ into the recipient. This shows that conventional WWTPs are inefficient in removing PFAS from wastewater and sludge, so advanced treatment techniques are needed.

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1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are persistent organic contaminants of anthropogenic origin that are considered very persistent, potentially bioaccumulative, and toxic (Wang et al., 2011a; Ahrens, 2011). There are currently >5000 PFAS on the global market (Chelcea et al., 2020), with new compounds being developed continually. PFAS are suitable for multiple applications, such as cookware, clothing, packaging material, electronics, cosmetics, and aqueous firefighting foams (Buck et al., 2011). PFAS can be released into the environment from diffuse pathways, e.g., atmospheric deposition and surface run-off (Ahrens and Bundschuh, 2014; Charbonnet et al., 2021), and other pathways, e.g., landfills (Busch et al., 2010; Benskin et al., 2012), wastewater treatment plants (WWTPs) (Jacob et al., 2021; Golovko et al., 2021), and firefighter training facilities (Ahrens et al., 2015; Hu et al., 2016). Consequently, PFAS are ubiquitous in the environment, even in sparsely populated areas (MacInnis et al., 2019; Skaar et al., 2019). Human exposure mainly occurs via consumption of contaminated food (Gebink et al., 2015) or drinking water (Gyllenhammar et al., 2019), inhalation of air and dust (Haug et al., 2011), use of PFAS-containing products (e.g., food packaging), and personal care products (Thépaut et al., 2021). Some PFAS, particularly long-chain PFAS such as PFOS (perfluorooctane sulfonate) and PFOA (perfluorooctanoate), have been banned or restricted since the late 2000s, leading to changes in production and exposure to short-chain PFAS and other alternative PFAS (Wang et al., 2017; Gomis et al., 2015). However, most PFAS are not regulated and knowledge about their sources, transport, fate, and exposure is scarce.

WWTPs are a major source of PFAS in aqueous environments (Golovko et al., 2021; Dauchy et al., 2017), but conventional WWTPs are not designed to remove organic micropollutants such as PFAS (Schuricht et al., 2017). In general, domestic and industrial wastewater show differing PFAS patterns, with typically higher PFAS concentrations in industrial wastewater than municipal wastewater (Hu et al., 2016; Bossi et al., 2008; Becker et al., 2008; Kim et al., 2021). Due to the widespread use and persistent properties of PFAS, wastewater influent concentrations can be influenced by households and catchment-specific point sources (e.g., industry, airports, military bases, landfills) (Nguyen et al., 2022a). PFAS removal in WWTPs has been shown to be related to influent concentrations (Kim et al., 2012), treatment processes (Schuricht et al., 2017), PFAS functional group and perfluorocarbon chain-length (Gagliano et al., 2020), and proportion of industrial wastewater (Becker et al., 2008). Further, PFAS formation through precursor transformation is common within WWTP processes, leading to elevated concentrations of perfluoroalkyl sulfonates (PFSA) and perfluoroalkyl carboxylates (PFCA) in the effluent (Houtz et al., 2016). Emissions to the atmosphere (Ahrens et al., 2011) and use of PFAS-contaminated sludge as fertilizer (Sepulvado et al., 2011) are other distribution pathways of PFAS into the environment. PFAS in WWTP effluent and recipient water have been extensively studied (Golovko et al., 2021; Dauchy et al., 2017; Arvaniti and Stasinakis, 2015; Kim et al., 2021), but PFAS mass flows in wastewater network systems and within WWTPs have received less attention (Gallen et al., 2018; Lenka et al., 2021).

The aim of this study was to determine mass flows of 26 PFAS in a wastewater network and within a WWTP. Specific objectives were to i) evaluate sources and fate of PFAS in a wastewater network, ii) determine the fate of PFAS within a WWTP, and iii) compare mass flows of PFAS within the wastewater system. The mass flows focused on the dissolved phase (filtered water samples) and sludge.

2. Materials and methods

2.1. Chemicals

In total 26 PFAS were investigated, comprising PFCA with

perfluorocarbon chain lengths C₃–C₁₃, C₁₅, and C₁₇ (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA, PFTeDA, PFHxDA, PFOcDA), C₄, C₆, C₈, and C₁₀ PFSA (PFBS, PFHxS, PFOS, PFDS), and the PFAS precursors methyl- and ethyl-perfluorooctanesulfonamides (FOSAs; FOSA, MeFOSA, EtFOSA), methyl- and ethylperfluorooctanesulfonamidoethanols (FOSEs; MeFOSE, EtFOSE), methyl- and ethylperfluorooctanesulfonamidoacetic acid (FOSAA; FOSAA, MeFOSAA, EtFOSAA), and 6:2 FTSA (Wellington Laboratories). Sixteen mass-labelled internal standards (ISs) were used in the analytical method: ¹³C₄-PFBA, ¹³C₂-PFHxA, ¹³C₄-PFOA, ¹³C₅-PFNA, ¹³C₂-PFDA, ¹³C₂-PFUnDA, ¹³C₂-PFDoDA, ¹⁸O₂-PFHxS, ¹³C₄-PFOS, ¹³C₈-FOSA, d₃-MeFOSA, d₅-EtFOSA, d₇-MeFOSE, d₉-EtFOSE, d₃-MeFOSAA, and d₅-EtFOSAA. Full names, CAS-registry numbers, supplier, and purity are shown in Table S1 in Supporting Information (SI).

2.2. Description of sampling sites and sample collection

The wastewater network and main WWTP in Uppsala, Sweden, were selected as study objects. The network consists of 52 pumping stations (PST), which receive domestic and industrial wastewater and some stormwater from Uppsala, Bälänge, and Lövsälöt communities. In 2016, the WWTP treated wastewater from 200,000 person-equivalents, corresponding to 19 million m³ wastewater (~52,800 m³ d⁻¹). Retention time in the WWTP is ~20 h. It started operating in 1945 and was expanded in 1957 (WWTP section A), 1967 (section B), and 1999 (section C) (Fig. 1). The treatment process comprises: i) primary mechanical treatment with grids, screening, aerated sand traps, and primary sedimentation with pre-precipitation of ferric chloride (FeCl₃) addition, ii) biological treatment with activated sludge in aerated and anoxic basins, and iii) chemical treatment with FeCl₃. Section A and B influents are dominated by municipal wastewater, while section C receives a mixture of domestic, industrial, and other commercial wastewater. The sludge (primary, waste activated sludge (WAS), chemical sludge) is thickened, anaerobically digested for biogas production (2.7 normalized million m³ in 2021) and dewatered (by centrifuges), creating a sludge product used as fertilizer or deposited on landfill. Primary sludge is treated in series in digesters 1 and 2, respectively, and activated sludge is treated only in digester 2. The treated wastewater is discharged into the Fyris River, which flows into Lake Mälaren, the largest drinking water reservoir in Sweden (serving ~2 million people).

Wastewater and sludge samples were collected in January 2016 (Table S2 in SI). In total, 35 wastewater samples (1 L) were collected, 15 from the wastewater PSTs in Uppsala and 20 from the WWTP. Two samples collected at the PSTs were time-integrated samples (24 h) and the others (*n* = 13) were grab samples, while at the WWTP nine samples were 24-h flow proportional samples and the others (*n* = 11) grab samples. Samples were also collected from sludge (*n* = 10) and reject water from sludge dewatering as a grab sample (*n* = 1) within the WWTP. Water samples were stored refrigerated (4.5 °C) and sludge samples were stored frozen (-20 °C) before analysis. Physical properties of the sludge (dry weight (dw), moisture content, organic matter (OM), density) are shown in Tables S3 and S4 in SI.

2.3. Sample preparation and analysis

Wastewater samples were extracted by solid-phase extraction (SPE) as described previously (McCleaf et al., 2017). Before extraction, the samples were sonicated for 5 min, filtered through glass fiber filters (GFF; 1.2 µm; GE Healthcare Life Sciences, Whatman, U.K.), and spiked with 100 µL of an internal standard (IS) mixture (*c* = 200 ng mL⁻¹ per compound). A 300 mL aliquot was then extracted using Oasis WAX cartridges (6 cm³, 500 mg, 60 µm, Waters Corporation, USA) pre-conditioned with 4 mL 0.1% ammonium hydroxide in methanol, 4 mL methanol, and 4 mL Millipore water. The flow rate through the cartridge was adjusted to ~1 drop s⁻¹. Cartridges were washed with 4 mL 25 mM ammonium acetate buffer in Millipore water and dried by centrifugation

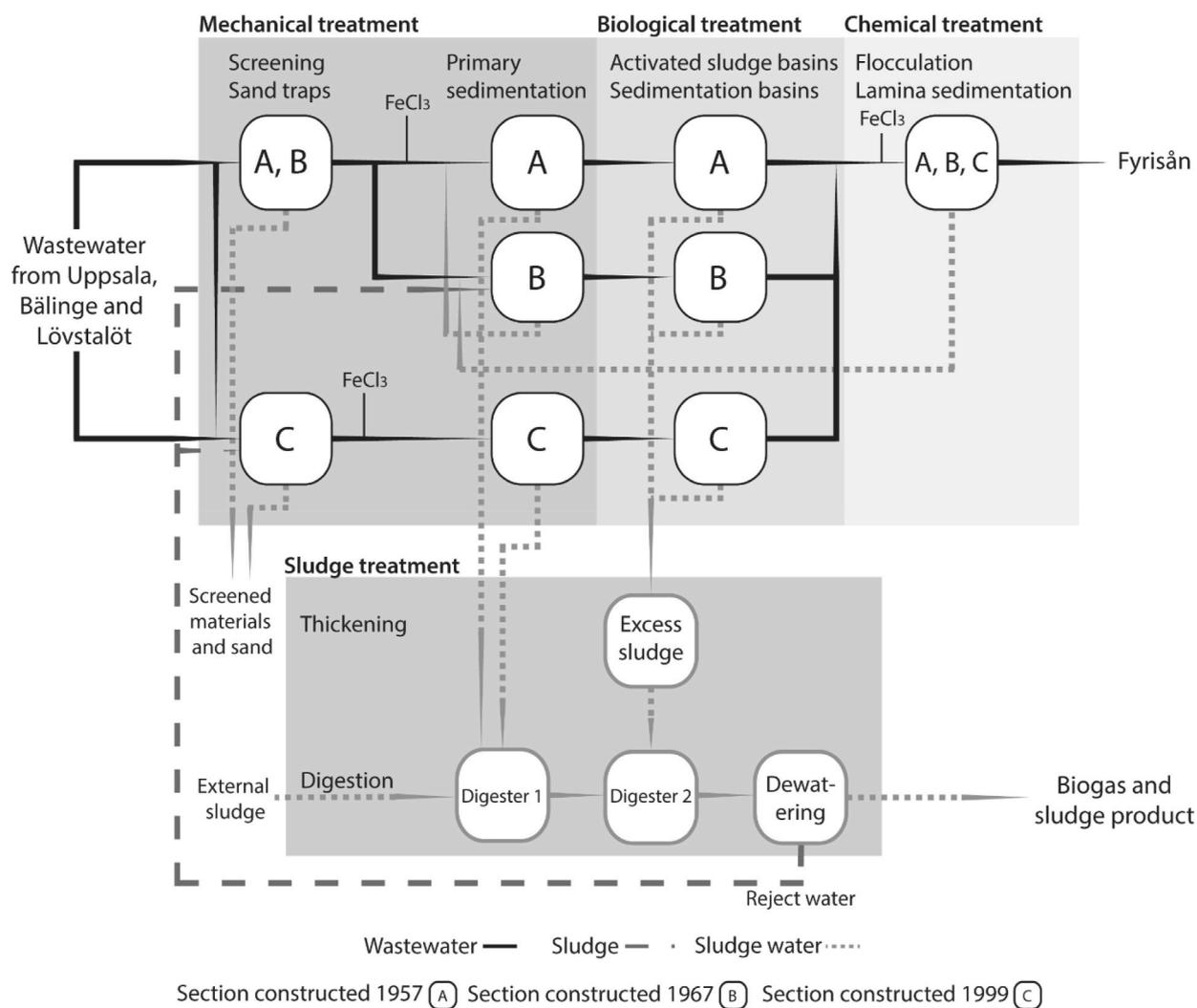


Fig. 1. Schematic diagram of Uppsala WWTP showing the different treatment sections for wastewater and sludge.

(3000 rpm, 2 min). The extracts were eluted with 4 mL methanol and 8 mL 0.1% ammonium hydroxide solution in methanol, and gently concentrated to 1 mL under a nitrogen stream at room temperature. The samples were analyzed using high-performance liquid chromatography-tandem mass spectrometry (HPLC-MS/MS) (Agilent Technologies 1200 Series, Palo Alto, CA, USA, and Agilent 6460 Triple Quadrupole System, Palo Alto, CA, USA). PFAS were quantified using the isotope dilution method.

Sludge samples were extracted by solid-liquid extraction (SLE) as described by Powley et al. (2005). Prior to extraction, samples were freeze-dried for seven days, homogenized (Bertin Technologies, Precellys Evolution), and 1–2 g portions were transferred to 50 mL PP tubes, 2 mL 100 mM sodium hydroxide in 80%/20% methanol/Millipore water were added, and the samples were left to soak for 30 min. Then 20 mL methanol and 100 μ L IS mixture were added, mixed on a wrist-action shaker (Gerhardt, Germany; 200 rpm for 60 min), and centrifuged (3000 rpm for 15 min) to separate solid and liquid phase. Finally, the supernatant was transferred to a new 50 mL PP tube. This procedure was repeated twice more, first with 1 mL sodium hydroxide in 80%/20% methanol/Millipore water and then with 10 mL methanol, each time mixed by the wrist-action-shaker (200 rpm for 30 min). Finally, 0.1 mL 4 M hydrochloric acid (HCl) was added to the extract, shaken manually, and centrifuged (3000 rpm for 5 min). Aliquots of the extract (8.3 mL) were transferred to 15-mL PP tubes and concentrated gently under a nitrogen stream to 1 mL. During the following clean-up step with 25 mg

ENVI-carb (Supelclean; Supelco; bulk packaging, 100 $\text{m}^2 \text{g}^{-1}$; 120/400 mesh) and 50 μ L glacial acetic acid, sample extracts were vortexed, centrifuged (4000 rpm for 15 min), and transferred to amber glass vials. All samples were stored in the freezer until analysis using HPLC-MS/MS (McCleaf et al., 2017).

2.4. Quality assurance and quality control

Sample process blanks consisted of MilliQ water for wastewater ($n = 2$) and extraction without sample matrix for sludge ($n = 3$) were analyzed in the same way as the samples. Method detection limit (MDL) was calculated as mean blank concentration plus three standard deviations. If a PFAS was not detected in the blank samples, the lowest calibration point was used. The MDL in wastewater was 0.19 ng L^{-1} for all PFAS except PFDA (3.9 ng L^{-1}), while the MDL in sludge ranged from 0.041 (6:2 FTSA) to 1.3 $\text{ng g}^{-1} \text{dw}$ (PFBA) (Table S5 in SI). Mean recovery was 100 \pm 39% in wastewater and 84 \pm 46% in sludge (Table S6 in SI). Mean relative standard deviation of triplicate sludge and duplicate water samples was 30% and 11%, respectively.

2.5. Data evaluation

Wastewater and sludge mass flows (mg d^{-1}) were calculated by multiplying the concentration measured at the PST or WWTP by discharge:

$$\text{Mass flow (mg d}^{-1}\text{)} = \text{Concentration (ng L}^{-1}\text{)} \bullet \text{Discharge (m}^3\text{d}^{-1}\text{)} \bullet 10^{-3} \tag{1}$$

Removal efficiency (RE) was calculated based on mass flow, considering the difference between influent and effluent:

$$\text{RE (\%)} = \frac{\text{Influent AB} + \text{Influent C} - \text{Effluent}}{\text{Influent AB} + \text{Influent C}} \bullet 100 \tag{2}$$

Mass flow of PFAS in sludge was calculated as:

$$\text{Mass flow (mg d}^{-1}\text{)} = \text{Concentration (ng g}^{-1}\text{ ww)} \bullet \rho \text{ (g mL}^{-1}\text{)} \bullet V \text{ (m}^3\text{ d}^{-1}\text{)} \tag{3}$$

where ρ is density of the sludge (g mL⁻¹) and V is sludge flow (m³ d⁻¹). The non-detect concentrations were handled as zero in the calculation of the \sum_{26} PFAS.

3. Results and discussion

3.1. PFAS concentrations, composition, and mass flow in the wastewater network

Of 26 PFAS analyzed, 21 (C₄, C₆ and C₈ PFSA, C₃–C₁₇ PFCA, FOSA, EtFOSA, MeFOSAA, EtFOSAA, 6:2 FTSA) were detected within the wastewater network (Fig. 2, Tables S7 and S8 in SI). The aqueous phase was filtered before extraction. Thus, the samples represent the dissolved phase as the PFAS sorbed to particulates would have been filtered out of the samples before analysis. \sum_{26} PFAS concentration ranged from 89 ng L⁻¹ (PST16) to 55,000 ng L⁻¹ (PST4), with PSTs receiving wastewater from mixed (domestic, industrial, commercial) sources showing the highest concentrations (mean 19,000 ng L⁻¹; median 180 ng L⁻¹), followed by industrial wastewater PSTs (mean 1000 ng L⁻¹; median 730 ng L⁻¹) and domestic wastewater PSTs (mean 290 ng L⁻¹; median 150 ng L⁻¹) (Table S9 in SI). However, some PSTs receiving domestic

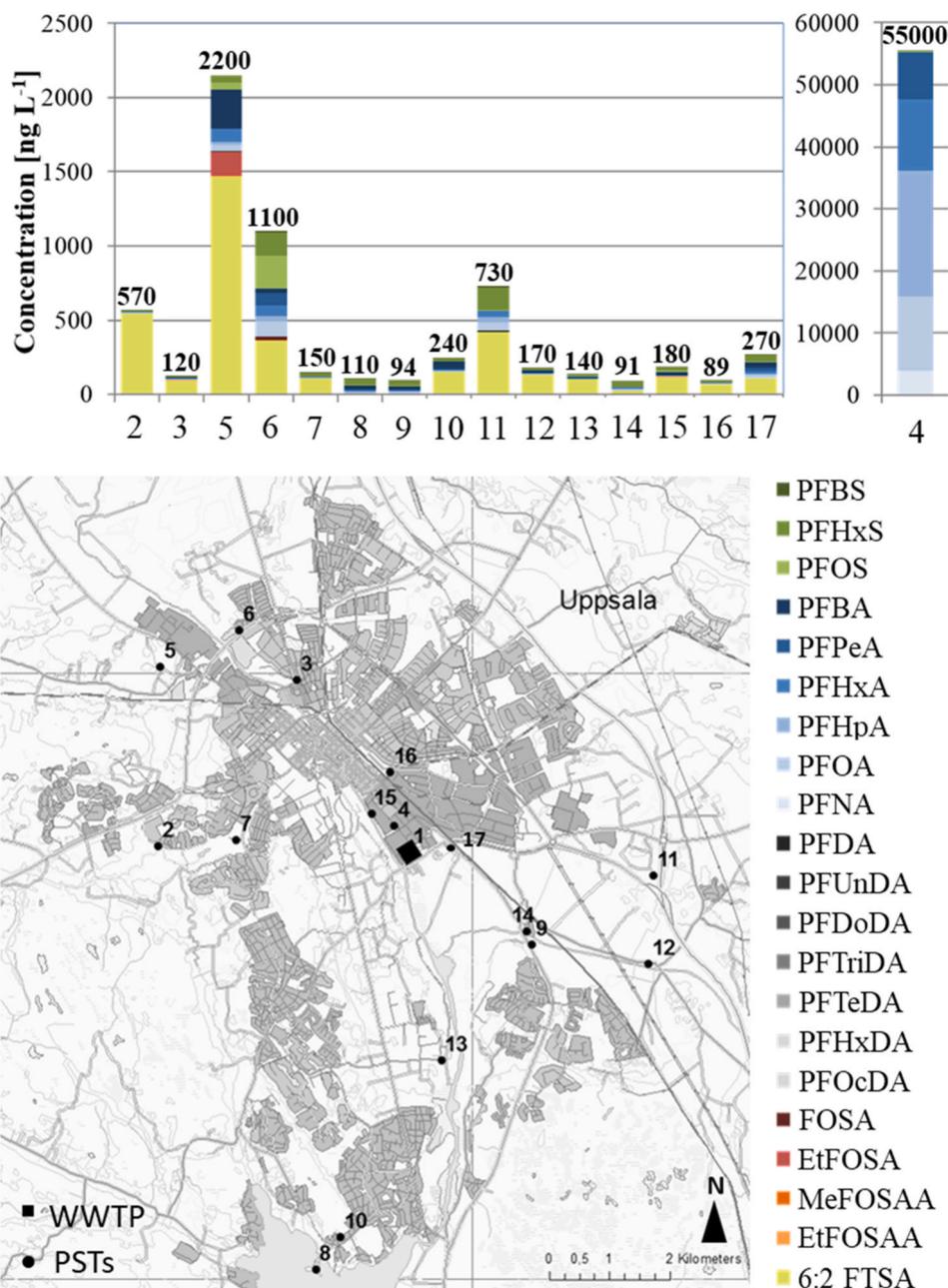


Fig. 2. Individual PFAS concentrations at the 15 pumping stations in Uppsala, Sweden. Values on bars (upper panel) are \sum_{26} PFAS concentration.

wastewater had higher \sum_{26} PFAS concentrations (e.g., PST6: 1100 ng L⁻¹ and PST2: 570 ng L⁻¹) than those receiving industrial or mixed wastewater (e.g., PST3: 120 ng L⁻¹, PST16: 90 ng L⁻¹, PST15: 180 ng L⁻¹). This indicates that industrial and commercial activities were not necessarily associated with release of PFAS to wastewater. However, PST4 was strongly impacted by industry and showed several orders of magnitude higher \sum_{26} PFAS concentrations than the other PSTs (55,000 ng L⁻¹), with a very distinct PFAS composition profile dominated by PFCA (99.7% of \sum_{26} PFAS), mainly PFHpA (36%), PFOA (22%), PFHxA (21%), and PFPeA (14%). It has been shown previously that PFOA emissions are highly correlated with the paper industry and that PFNA, PFDA, and PFDoDA are mainly emitted from industrial sources (Clara et al., 2008; Guo et al., 2010; Barisci and Suri, 2021).

6:2 FTSA was the dominant PFAS (detected at all PSTs except PST4), representing 40–96% of PFAS (mean concentration 230 ng L⁻¹; median

150 ng L⁻¹). Elevated 6:2 FTSA levels at PST6 and PST11 might originate from a nearby firefighter training facility, which is highly contaminated with PFAS (Gyllenhammar et al., 2015, 2019). However, there is a lack of studies on 6:2 FTSA in wastewater networks (Arvaniti and Stasinakis, 2015). Other dominant PFAS in the PSTs were: PFHxS (14% of \sum_{26} PFAS), PFBA (8%), and PFHxA (7%). PST6, which receives domestic wastewater and runoff from a PFAS-contaminated site, had the highest PFOS concentration (220 ng L⁻¹). According to previous studies, PFOS is more dominant in domestic wastewater (Chen et al., 2012), while PFOA often dominates in industrial wastewater (Kim et al., 2012). This was not confirmed in this study, possibly because of phase-out of PFOS and a subsequent shift to short-chain and other PFAS (Wang et al., 2017). Short-chain PFCA were frequently detected, with C₅–C₇ PFCA occurring at all PSTs. C₉–C₁₇ PFCA were less frequently detected, but were found at several PSTs receiving industrial (n = 3) or domestic

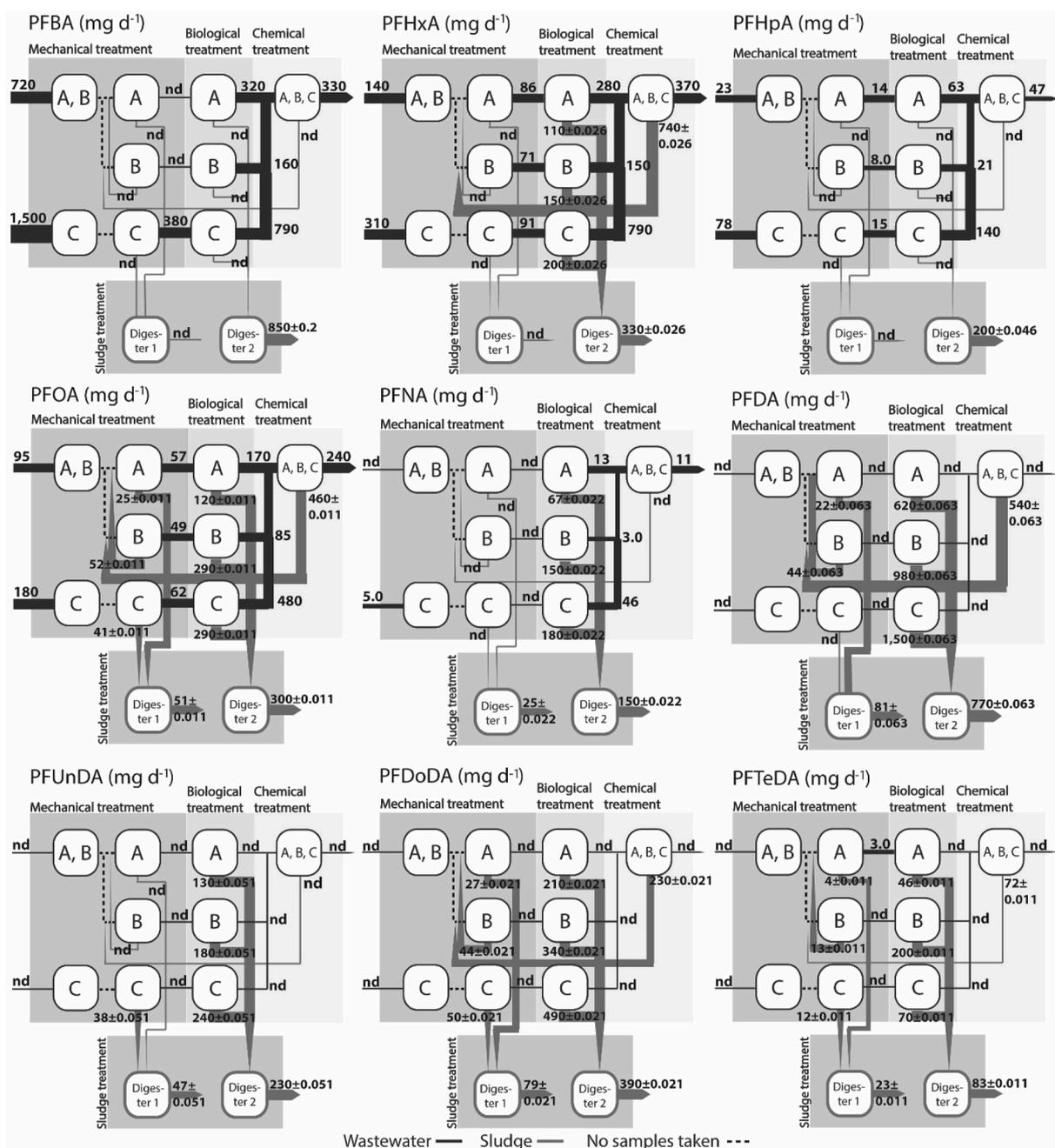


Fig. 3. Mass flows (mg d⁻¹) of PFCA in wastewater and sludge at different sampling points (influent and between different treatment steps) in the Uppsala WWTP. Influent A and B are dominated by municipal wastewater, while C receives a mixture of domestic, industrial, and commercial wastewater.

wastewater ($n = 5$), where they represented 0.011–1.1% of \sum_{26} PFAS. This study focused on the dissolved phase and the longer chained C_9 – C_{17} PFCA were likely removed during the filtration process since they are predominantly bound to the particulate phase (Arvaniti et al., 2014).

PFAS detected in >50% of the PST samples ($n = 10$ PFAS) showed significant intercorrelations (Pearson correlation coefficient; Table S10 in SI). PFOS was significantly correlated with PFCA (e.g., PFHxA, PFHpA, PFOA, PFNA) and C_5 – C_8 PFCA were correlated with each other ($p < 0.0001$), indicating related sources. However, 6:2 FTSA was significantly negatively correlated with C_5 – C_8 PFCA, which may indicate that it is used as a replacement for PFCA.

The highest mass flow of \sum_{26} PFAS was detected at PST4 (\sum_{26} PFAS = 110 g d^{-1}), 26-fold higher than the second highest (PST15: 4.3 g d^{-1}), and 72,000-fold higher than the lowest mass flow (PST7: 1.5 mg d^{-1}) (Table S11 in SI). Comparing the different PFAS classes, \sum PFCA had the highest mass flow at PST4 (106 g d^{-1} , dominated by PFHpA at 39 g d^{-1}), which was also the highest individual mass flow within the network. For the PFAS precursors, 6:2 FTSA was dominant, with \sum PSTs mass flow of 5 g d^{-1} . \sum PFSA had the lowest mass flow, ranging from 0.29 mg d^{-1} (PST7) to 730 mg d^{-1} (PST15), dominated by PFOS (\sum PSTs = 690 mg d^{-1}), followed by PFHxS (\sum PSTs = 680 mg d^{-1}) and PFBS (\sum PSTs = 130 mg d^{-1}), while PFDS was not detected at all.

3.2. PFAS in the wastewater at the WWTP

Within the WWTP, 13 of 26 PFAS analyzed were detected in wastewater (C_4 , C_6 , C_8 PFSA, C_3 – C_8 , C_{14} PFCA, FOSA, EtFOSAA, 6:2 FTSA). Among individual PFAS, 6:2 FTSA had the highest concentration (190 ng L^{-1} , median 110 ng L^{-1}) in WWTP wastewater, followed by PFHxS (max: 82 ng L^{-1} , median: 19 ng L^{-1}), and PFBA (max: 75 ng L^{-1} , median: 15 ng L^{-1}) (Figs. 3–5). The composition profile was dominated by 6:2 FTSA (27–73%; Fig. 4) in all WWTP treatment steps, followed by PFHxS (5–18%; Fig. 4) and PFBA (0–36%; Fig. 3). In influent A/B (mainly domestic wastewater), 6:2 FTSA (58%) dominated, while in influent C (mixture of domestic, industrial, and commercial wastewater), C_3 – C_8 PFCA dominated, with 37% PFBA. In WWTP effluent, 6:2 FTSA (72%) again dominated, followed by PFHxS (8%). The long-chain C_9 – C_{17} PFCA, except for PFTeDA, were not detected in wastewater, presumably due to high sorption to sludge (reported in a later section). PFHxS increased in all treatment steps except mechanical treatment. However, the effluent concentration of PFHxS (14 ng L^{-1}) was lower than the influent concentration (19 and 34 ng L^{-1} for influents A/B and C, respectively) (Fig. 4), indicating loss to the sludge (Arvaniti et al., 2014).

Ten of the 13 PFAS detected in the WWTP were also detected in the influent, while PFPeA and PFTeDA were only detected after the primary

clarifier (mechanical treatment) and FOSA only after the secondary clarifier (biological treatment). Detection of these PFAS within the WWTP, but not in influent, indicates transformation of PFAS precursors to PFCA and FOSA, as observed in other studies (Houtz et al., 2016; Eriksson et al., 2017) (Figs. 3–5). EtFOSAA was detected in influent but not in effluent, probably due to transformation during the WWTP treatment process (Rhoads et al., 2008). Overall, \sum_{26} PFAS concentration in WWTP effluent was slightly lower (170 ng L^{-1}) than in influent (site A/B = 190 ng L^{-1} ; site C = 210 ng L^{-1}) due to adsorption to sludge (Schuricht et al., 2017) or transformation of PFAS precursors (Houtz et al., 2016). The highest PFAS concentration was detected after secondary clarifier A (biological treatment) (\sum_{26} PFAS = 440 ng L^{-1}), dominated by PFSA (34% of \sum_{26} PFAS), PFCA (33%), and PFAS precursors (33%), indicating presence of a variety of PFAS (Arvaniti and Stasinakis, 2015). Most PFAS concentrations in the wastewater increased after biological treatment, in particular for short-chain PFCA and sometimes PFSA, while some PFAS concentrations decreased (PFTeDA, EtFOSAA). Increasing concentrations after biological treatment have been reported previously for PFOS (Schultz et al., 2006; Becker et al., 2008) and can be explained by transformation of PFAS precursors to PFCA and PFSA and formation of short-chain PFAS during aerobic and anaerobic biological processes (Rhoads et al., 2008; Arvaniti et al., 2014; Gagliano et al., 2020).

Calculated mass flows in the WWTP were highest for the secondary clarifier C (\sum_{26} PFAS = 7700 mg d^{-1}), followed by effluent (\sum_{26} PFAS = 7000 mg d^{-1}) and influent (C: \sum_{26} PFAS = 4200 mg d^{-1}). 6:2 FTSA had the highest mass flow in influent (AB: 2200 mg d^{-1}) and effluent (5000 mg d^{-1}), followed by PFBA (1500 mg d^{-1} , influent C) and PFHxS (AB = 380 mg d^{-1} and C = 690 mg d^{-1} ; effluent = 560 mg d^{-1}). These differences are likely attributable to sorption and desorption from sludge recirculating in the WWTP and to transformation of PFAS precursors (Arvaniti et al., 2014; Rhoads et al., 2008). Schultz et al. (2006) calculated mass flows of 0.14 – 0.26 g d^{-1} for 6:2 FTSA in a WWTP (influent and effluent) in the US, which is around one order of magnitude lower than our values (1.1 – 5.0 g d^{-1}). Effluent mass flows for PFOA (0.29 – 0.40 g d^{-1}) and PFOS (0.40 – 0.64 g d^{-1}) in the same plant were in the same range as in our study. In contrast, Sinclair and Kannan (2006) recorded higher effluent mass flows of PFOA (0.38 – 32 g d^{-1}) in industrially influenced WWTPs in the US, while PFOS effluent mass flows (0.012 – 4.1 g d^{-1}) were similar to ours. A study in Bavaria, Germany, observed rather high effluent mass flows, on average 2.8 g d^{-1} for PFOS and 2.2 g d^{-1} for PFOA (Becker et al., 2008). Differences between studies are likely due to wastewater origin, WWTP steps, and the relatively recent regulation of certain PFAS (Sinclair and Kannan, 2006; Becker et al., 2008; Schuricht et al., 2017). Per capita discharge in this study was estimated to be $43 \mu\text{g day}^{-1}$ for \sum_{26} PFAS, $31 \mu\text{g day}^{-1}$ for 6:2 FTSA,

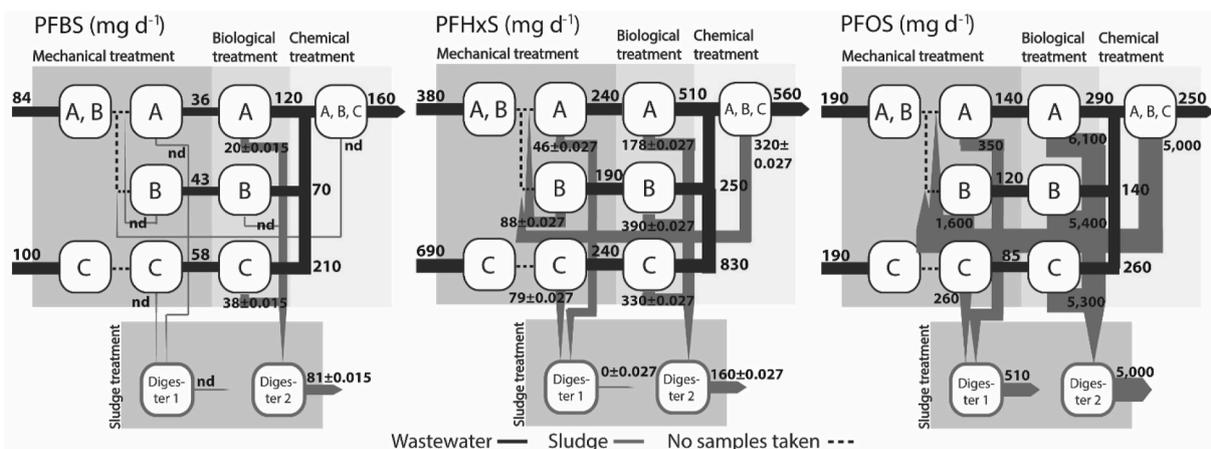


Fig. 4. Mass flows (mg d^{-1}) of PFASs in wastewater and sludge at different sampling points (influent and between different treatment steps) in the Uppsala WWTP. Influent A and B are dominated by domestic wastewater while C receives a mixture of domestic, industrial, and commercial wastewater.

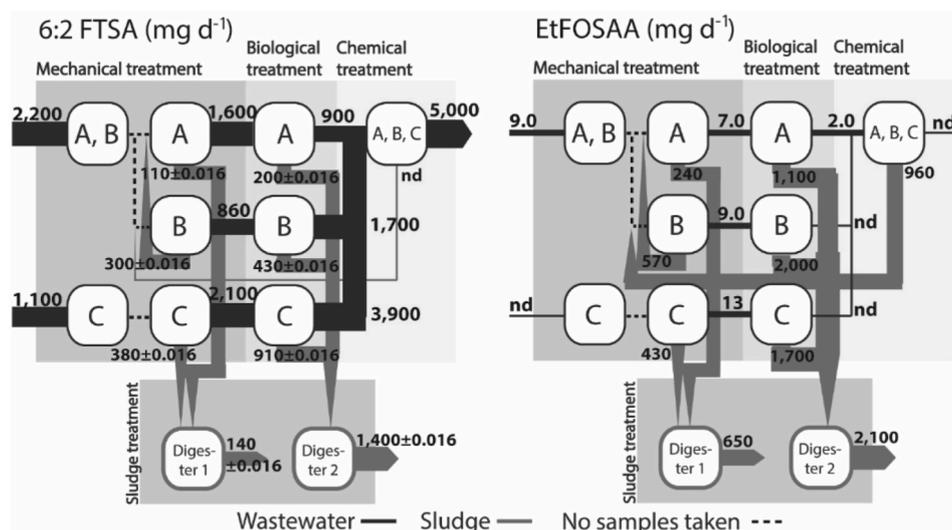


Fig. 5. Mass flows (mg d^{-1}) of the PFAS precursors 6:2 FTSA and EtFOSAA in wastewater and sludge at different sampling points (influent and between different treatment steps) in the Uppsala WWTP. Influent A and B are dominated by domestic wastewater while C receives a mixture of domestic, industrial and commercial wastewater.

$3.5 \mu\text{g day}^{-1}$ for PFHxS, $2.3 \mu\text{g day}^{-1}$ for PFHxA, $2.0 \mu\text{g day}^{-1}$ for PFBA, and $1.5 \mu\text{g day}^{-1}$ for PFOS and PFOA. These are within the range reported in studies in the US (Barisci and Suri, 2021), Australia (Nguyen et al., 2022a), Korea (Kim et al., 2012), and Belgium (Jeong et al., 2022).

Removal efficiency (based on dissolved wastewater concentrations) varied greatly among individual PFAS and within the WWTP, leading to mean removal efficiency of $10 \pm 68\%$ for individual PFAS. Concentrations of six PFAS ($\text{C}_4\text{--}\text{C}_8$ PFCA, 6:2 FTSA) slightly increased and those of five PFAS (PFBA, C_4 , C_6 , C_8 PFSA, EtFOSAA) slightly decreased. The slight increase in PFCA could be due to PFAS precursor degradation (Dauchy et al., 2017) and the decrease in PFSA and EtFOSAA could be due to sorption to sludge (Arvaniti et al., 2014) and degradation (EtFOSAA) (Rhoads et al., 2008). Since this study focused on the dissolved phase, the removal efficiency might be underestimated. However, the low removal efficiency of PFAS in the here investigated conventional WWTP is consistent with previous findings (Schultz et al., 2006; Bossi et al., 2008; Lenka et al., 2021). Ultimately, the WWTP effluent is discharged into the recipient Fyris River, which has shown elevated PFAS concentrations in the surface water downstream of the WWTP (mean $26 \pm 15 \text{ ng L}^{-1}$) compared to upstream of the WWTP ($11 \pm 5.0 \text{ ng L}^{-1}$) (Nguyen et al., 2022b).

3.3. PFAS in WWTP sludge

Of the 26 PFAS analyzed, 21 were detected in WWTP sludge (C_4 , C_6 , C_8 , and C_{10} PFSA, $\text{C}_3\text{--}\text{C}_{16}$ PFCA, EtFOSA, EtFOSAA, FOSAA, and MeFOSAA). Interestingly, some PFAS (PFHpA, FOSAA, MeFOSAA) detected in digested sludge from digester 1 have not been found previously in primary clarifier sludge (Table S8 in SI). A similar trend was observed for digested sludge from digester 2, where PFBA and PFHpA were only found in the digested sludge, while PFDS and PFPeA were only detected prior to digestion. These changes in PFAS composition could be due to varying PFAS concentrations over time, precursor transformation (Houtz et al., 2016), and sampling and analytical uncertainties. Most PFAS detected in sludge were also found in dewatered sludge ($n = 15$), and some even in the reject water from dewatering ($n = 5$). However, some PFAS in the reject water (PFBS, PFHpA) were not detected in the dewatered sludge, as these short-chain PFAS have higher water solubility than long-chain PFAS (Wang et al., 2011b). PFOA and PFDA had higher sludge concentration than PFNA and PFUnDA, respectively, indicating a preference of even-chain PFAS, but not odd-chain PFAS, for sludge partitioning, as observed previously (Bossi

et al., 2008). PFOS showed the highest concentrations (mean 13 ng g^{-1} dw; maximum 38 ng g^{-1} dw) of all PFAS analyzed (Fig. 4), and was the dominant compound in sludge (11–62% of $\sum_{26}\text{PFAS}$ concentration). Dominance of PFOS in sludge has been reported previously (Arvaniti et al., 2014). In primary clarifier A, primary clarifier C, and digested sludge from digester 1, the PFAS precursors EtFOSA, FOSAA, MeFOSAA, and EtFOSAA were the dominant group, representing 59–86% of $\sum_{26}\text{PFAS}$ (based on dw). In primary clarifier A, EtFOSA dominated (75% of $\sum_{26}\text{PFAS}$), likely due to transformation of EtFOSE to EtFOSAA during aerobic treatment (Rhoads et al., 2008).

The $\sum_{26}\text{PFAS}$ concentrations in dewatered (33 ng g^{-1} dw) and digested ($12\text{--}36 \text{ ng g}^{-1}$ dw) sludge were higher than those detected in primary clarifier sludge ($7.0\text{--}34 \text{ ng g}^{-1}$ dw). The highest $\sum_{26}\text{PFAS}$ concentration was found in secondary clarifier sludge (61 ng g^{-1} dw), and the lowest in primary clarifier sludge (7.0 ng g^{-1} dw). The 44–86% increase in PFAS concentrations from primary to secondary clarifier indicates high transfer/sorption of PFAS from the dissolved to the particulate phase during biological treatment (Arvaniti et al., 2014). This might have caused the 82% decrease in $\sum_{26}\text{PFAS}$ concentration in the final clarifier sludge (9.6 ng g^{-1} dw) compared with the secondary clarifier sludge (mean 54 ng g^{-1} dw), as most PFAS (particularly PFSA, $\text{C}_3\text{--}\text{C}_8$ PFCA, FOSAA, and 6:2 FTSA) had already been transferred to the particulate phase (Arvaniti et al., 2014).

Mass flows in the sludge varied for individual PFAS, with the highest $\sum_{26}\text{PFAS}$ mass flows from digested sludge (digester 2; $13,000 \text{ mg d}^{-1}$), followed by secondary clarifier B ($12,500 \text{ mg d}^{-1}$) and secondary clarifier C ($12,000 \text{ mg d}^{-1}$) sludge. PFOS and PFHxS showed the highest mass flows in sludge, 5000 mg d^{-1} (digester 2) and 390 mg d^{-1} (secondary clarifier B). The PFOS mass flow (mean 3300 mg d^{-1}) was more than 10-fold higher than the mean of 210 mg d^{-1} reported for a US municipal WWTP (Schultz et al., 2006). Increased mass flow after the secondary clarifier was found for PFBS, PFHxS, and PFOS, a trend also observed in the wastewater. All $\text{C}_3\text{--}\text{C}_8$ PFCA had high mass flows from digested sludge (digester 2), with PFBA having the highest (850 mg d^{-1}), even though it is a short-chain PFCA with higher water solubility that was expected to remain in the dissolved phase (i.e., wastewater) (Wang et al., 2011b). PFAS concentrations mostly decreased after the secondary clarifier, while PFHxA and PFOA showed an increase in mass flow and PFPeA was not detected after the secondary clarifier. $\text{C}_9\text{--}\text{C}_{17}$ PFCA were rarely detected in the wastewater, but showed high mass flows in sludge, with a maximum of 1500 mg d^{-1} for PFDA, 500 mg d^{-1} for PFDoDA, and 240 mg d^{-1} for PFUnDA. High mass flows of EtFOSA were

only detected in primary clarifier sludge A (2500 mg d⁻¹), but it had the highest mass flow of all PFAS precursors, while EtFOSAA mass flows in sludge increased after all secondary clarifiers (Fig. 3). Sludge is recirculated in the WWTP, so final clarifier sludge goes back to primary clarifier B, then continues to primary clarifier A. In general, PFAS concentrations were consistently high in all sludge samples across all treatment steps for PFHxS, PFOS, PFHxA, PFOA, EtFOSAA, and 6:2 FTSA, which can be explained by recirculation of sludge within the WWTP (Schultz et al., 2006). Recirculation of wastewater within the treatment plant could also explain the high concentrations and mass flows of EtFOSA in sludge from primary clarifier A. Other studies have shown that residence time in anaerobic digesters can cause variation in mass flows or degradation of precursors, resulting in increasing mass flow in digestate (Schultz et al., 2006). However, the PFAS precursors FOSAA and 6:2 FTSA do not show decreasing concentrations in any anaerobic digestion process, indicating that they are more resistant to anaerobic digestion than EtFOSA (Rhoads et al., 2008). Ultimately, sludge is an important sink within the WWTP and source of PFAS to the environment when applied as biosolids on agricultural land (Fredriksson et al., 2022; Lakshminarasimman et al., 2021).

4. Conclusions

Our findings on PFAS in the wastewater network system helped identify sources and mass transport processes. Highly elevated concentrations of C₃–C₈ PFCA at PST4 were likely caused by an industrial source connected to the municipal wastewater network system. Elevated concentrations of 6:2 FTSA at PST6 and PST11 may originate from a nearby PFAS-contaminated site. Within the WWTP, shorter-chain PFAS (C₃–C₇ PFCA, C₄, C₆ PFSA, 6:2 FTSA) dominated in wastewater, whereas longer-chain PFAS (C₈–C₁₃ PFCA, PFOS, EtFOSAA) dominated in sludge. Short-chain PFAS are more mobile and likely to be transported with the aqueous phase. Decreases in PFSA and EtFOSAA concentrations within the WWTP could be due to sorption to sludge, but also degradation (EtFOSAA). The consistently high PFAS concentration in sludge is likely attributable to sludge recirculation within the WWTP. Mass flow data on PFAS improved understanding of PFAS cycling within the WWTP, including sorption and desorption to particles and sludge, recirculation within the WWTP, and transformation of PFAS precursors. Overall, PFAS were not efficiently removed in the WWTP (mean removal efficiency 10 ± 68% for individual PFAS), resulting in discharge of 7000 mg ∑₂₆PFAS d⁻¹ into the recipient. Thus conventional municipal WWTPs are inefficient in removal of PFAS from wastewater and sludge, and control of upstream sources or advanced treatment techniques are needed.

Credit author statement

Laura Gobelius: Data curation, Writing- Original draft preparation, Validation, Formal analysis. **Linda Glimstedt:** Methodology, Data curation, Validation, Formal analysis. **Jesper Olsson:** Writing- Reviewing and Editing. **Karin Wiberg:** Writing- Reviewing and Editing, Validation. **Lutz Ahrens:** Project administration, Supervision, Conceptualization, Writing- Reviewing and Editing, Validation.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests. Lutz Ahrens reports financial support was provided by Uppsala Water and Waste AB, Sweden.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2023.139182>.

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