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Stormwater discharges affect PFAS occurrence, concentrations, and spatial distribution in water and bottom sediment of urban streams

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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are extensively used in urban environments and are, thus, found in urban stormwater. However, the relevance of stormwater as a pathway for PFAS to urban streams is largely unknown. This study evaluated the impact of urban stormwater runoff on PFAS concentrations and spatial distribution in three urban streams affected by stormwater discharges from separate sewer systems. River water was sampled during dry (DW) and wet weather (WW) upstream, immediately downstream, and further downstream of three urbanized areas with separate sewer systems and with and without point sources (i.e. waste water treatment plant, airports). Water samples were analyzed for 34 targeted PFAS compounds and sediment samples for 35 targeted PFAS and 30 PFAS compounds using a total oxidizable precursor assay. The sum of the quantified PFAS concentrations ranged from the reporting limit (RL) to 84.7 ng/L during DW and increased as the streams were affected by WW discharges (0.87 to 102.3 ng/L). The highest PFAS concentrations were found downstream of urban areas and/or point sources (i.e. airports) during WW, indicating a clear contribution from stormwater discharges. A consistent PFAS contribution from the WWTP was observed under both DW and WW conditions. During WW events, concentrations of perfluorooctanesulfonic acid (PFOS) and total PFAS (PFOA equivalents) exceeded the annual average environmental quality standards, which are an established limit of 0.65 ng/L for PFOS and a proposed limit of 4.4 ng/L for total PFAS. Notably, except for the legacy PFAS, PFOS and perfluorooctanoic acid (PFOA), the most frequently quantified PFAS during DW were short-chain. For WW, long-chain perfluorocarboxylic acids (PFCAs) and a precursor, 6:2 Fluorotelomer sulfonic acid (6:2 FTS), were more frequently quantified, suggesting stormwater is a source of these longer-chain and particle-associated PFAS. The detection of unregulated fluorotelomer sulfonates (FTSs) such as 6:2 and 8:2 FTS during WW suggests a need for regulatory action, as these compounds can degrade into more stable PFAS. In sediment, higher concentrations, and a greater variety of PFAS were found at sites with known point sources i.e. airports. Long-chain PFCAs (C7–C13), perfluoroalkyl sulfonates (PFSAs) (C6), and precursors (i.e. N-Ethyl perfluorooctane sulfonamidoacetic acid), were more prevalent in sediments than in the water. Notably, PFOS concentrations in sediment exceeded the lowest Predicted No-Effect Concentration (PNEC) across sites, posing a potential long-term environmental risk, though current PNECs for other PFAS may underestimate such risks. The findings of the study highlight urban stormwater as a source of PFAS to urban streams indicating the need to minimize PFAS sources in the urban environment and to effectively treat stormwater to protect receiving water bodies.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a group of synthetic chemicals used in various industrial processes and consumer products, including aqueous fire-fighting foams (AFFFs), lubricants, aviation hydraulic fluids, personal care products, coatings, and paints (Glüge et al., 2020; Kurwadkar et al., 2022). The strong carbon-fluorine (C—F) bonds in PFAS provide high thermal and chemical stability (Key et al., 1997). The polar head and non-polar tail of these molecules make them both hydrophilic and hydrophobic (Moody and Field, 2000). While these characteristics make PFAS valuable for industrial applications, they also present environmental challenges. Due to their persistence, they are resistant to degradation (Buck et al., 2011). Once in the environment, they remain highly mobile, potentially bio-accumulative and toxic

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(Buck et al., 2011; Conder et al., 2008; Podder et al., 2021; Stahl et al., 2011).

PFAS have been widely used since the 1940s, and are present in surface waters due to contamination from wastewater treatment plants (WWTP), landfill sites, manufacturing emissions, and sites where AFFFs are used e.g. airports (Kurwadkar et al., 2022). PFAS have also been found in stormwater from various urban settings: streets, roads, other trafficked (Gasperi et al., 2022; Kim and Kannan, 2007; Murakami et al., 2009; Xiao et al., 2012), residential (Kim and Kannan, 2007; Xiao et al., 2012), industrial, and commercial (Xiao et al., 2012) areas. Here, possible sources may include atmospheric deposition (Kim and Kannan, 2007; Xiao et al., 2012), building materials (Bečanová et al., 2016; Liu et al., 2024), AFFFs (D'Agostino and Mabury, 2014), traffic-related sources (i.e. hydraulic fluids, lubricant oils) (Zhu and Kannan, 2020), as well as various consumer products (e.g. textile fabrics, food packing material) (Glüge et al., 2020). Since stormwater is often discharged into water bodies largely without treatment, it potentially transports PFAS to streams passing through urban areas. Given the presence, recurrence, persistence, mobility, and fate of PFAS in surface waters (Podder et al., 2021), it is particularly important to understand the role of stormwater runoff plays in carrying PFAS from urban areas to streams. Although several studies have been carried out on urban streams (Chen et al., 2017; Meyer et al., 2011; Müller et al., 2011; Nguyen et al., 2022, 2011; Sörengård et al., 2022; Zushi et al., 2008; Zushi and Masunaga, 2009), only a few have specifically investigated changes in urban streams resulting from rainfall-runoff events (Meyer et al., 2011; Müller et al., 2011; Nguyen et al., 2011; Zushi et al., 2008; Zushi and Masunaga, 2009). Further, most of these studies focused on a single watercourse (Meyer et al., 2011; Müller et al., 2011; Zushi et al., 2008; Zushi and Masunaga, 2009) with one (Meyer et al., 2011; Müller et al., 2011) or two (Zushi and Masunaga, 2009) events. The large variation of stormwater quality between events and catchments limits generalization of these findings, making it difficult to determine the significance of stormwater runoff as a source of PFAS pollution to the receiving water bodies. Additionally, previous studies have primarily focused on perfluorocarboxylic acids (PFCAs) and perfluoroalkyl sulfonates (PFSAs) only, thus leaving PFAS precursors unaddressed, and have not examined water-sediment interactions (Meyer et al., 2011; Müller et al., 2011; Nguyen et al., 2011; Zushi and Masunaga, 2009).

To understand the occurrence, concentration, and distribution of PFAS in urban streams, comprehensive studies are needed, including analyses of PFCAs, PFSAs, and precursors (e.g. fluorotelomer sulfonates). Understanding the extent to which stormwater runoff carries these pollutants from urban areas is essential for identifying the environmental risks associated with PFAS in stormwater runoff. Further, sediments can act as both sinks and secondary sources of PFAS (Ahrens et al., 2010; Chen et al., 2017). Thus, understanding the distribution of PFAS in sediment in urban streams is particularly important for predicting their long-term environmental impact, bioavailability, and ecological risks. Therefore, the purpose of this study was i) to investigate the occurrence of PFAS and their precursors in three urban streams with distinct characteristics, (ii) to investigate the potential contribution of stormwater runoff to PFAS by comparing DW and WW flows across various DW and WW events, (iii) to analyze how PFAS concentrations change along the watercourse from rural upstream to urbanized downstream areas, and (iv) to investigate the distribution of PFAS in water and sediment in urban streams.

2. Materials and methods

2.1. Presentation of sampling sites

Water and sediment samples were collected along three Swedish streams passing through urban areas: Fyrisån in Uppsala (Upp), Ljurabäck in Norrköping (Nrk), and Storån in Söderköping (Sdk). Each city has distinctive characteristics (Tables S1–S6) and operates separate sewer systems. Consequently, during WW events, the water bodies receive stormwater discharges from multiple outlets, typically without any prior treatment.

Three sampling locations were set along each stream (Table S7, Fig. S1). Upstream sampling sites, abbreviated to NrkA, SdkA, and UppA, acted as reference sites unaffected by urban runoff. The second locations, abbreviated to NrkB, SdkB, and UppB1, were located directly downstream of most of the urban area to evaluate the possible impact of urbanization and stormwater runoff. Uppsala is the only city with a WWTP discharging into the stream. For this reason, an additional sampling site, UppB2, was added approximately 100 m downstream from the WWTP discharge point and 200 m downstream from UppB1. The last locations, NrkC, SdkC, and UppC, were placed further downstream to monitor potential dilution effects; approximately 2 km downstream of sampling sites B (Table S7). Although B and C sampling sites were similarly spaced across streams, the C sites' proximity to urban areas varied. In Söderköping, the C site was far from the city center, while in Norrköping, it was close to an industrial area due to the stream's short length. While the upstream locations were intended as reference sites, the UppA sampling site, despite the absence of urban areas, is subject to stormwater and groundwater impact from a military air -force base which was a known PFAS source to the water body due to the use of AFFF (Gyllenhammar et al., 2015). The air force base discharges its stormwater into the stream through multiple outlets, both upstream and downstream of the UppA location. Similarly, part of the stream in Norrköping is impacted by a civil airport, also a known PFAS source, which discharges its stormwater into the stream through two outlets located between the NrkB and NrkC sampling sites. The potential groundwater impact of these airports on the streams remains unclear.

2.2. Sampling procedure

For water sampling, each sampling site was equipped with an autosampler (Teledyne ISCO 6712) with eight 1.8 L glass bottles to collect time-proportional sub-samples from the streams, approximately 0.5 m below the water surface (International Organization for Standardization, 2014), during both DW and WW conditions. DW was defined as days with less than 1 mm of precipitation, representing the concentration of compounds without direct input from stormwater runoff. WW events were defined as periods with more than 3 mm of precipitation or snowmelt, with samples collected during these events reflecting stream concentrations influenced by runoff events. The samplers were programmed to operate at time-proportional intervals based on the predicted duration of WW events. Sampling continued for 1 to 2 h after each WW event to capture all runoff from the contributing catchments to the streams. The duration of the WW events ranged from 3 to 96 h, while DW sampling was consistently carried out for 24 h. During DW sampling, subsamples (300 mL each) were taken every hour during a 24 h period resulting in a 7.2 L composite sample. For WW sampling, subsample volumes ranged from 180 mL to 360 mL each, with the sampling frequency varying from 15 min to 4 h, depending on the duration of the WW event (Table S8). The subsamples were retrieved within 24 h after the campaigns and thoroughly mixed in the field to create the 7.2 L composite sample which was distributed to the containers using a glass funnel, transported to the accredited laboratory in cooled bags, and kept in the dark at approximately 0–5 °C during transportation.

Sampling took place between June 2022 and October 2023, covering six WW ($n = 6$) and four DW events ($n = 4$) in Uppsala, seven WW ($n = 7$, of which one snowmelt event) and three DW events ($n = 3$) in Norrköping, and eight WW ($n = 8$, of which one snowmelt event) and three DW events ($n = 3$) in Söderköping (Table S8). Antecedent dry days ranged from 0 to 31, providing diverse conditions (Table S8).

As sediment quality is more stable than water quality, one sediment sampling event was carried out in September 2023 as close as possible to the water sampling locations previously described (Table S7). For sample collection, a powder-coated brass Ekman sediment sampler

(Hydro-bios) was used. At each site, several grab samples were taken and composited. Initially, these grab samples were collected in a stainless steel 9 L bucket, thoroughly mixed and then transferred to stainless steel trays. Due to the high -water content of the sediment, the coning and quartering method was not feasible. Thus, a mixing technique for homogenization adopted from Flanagan et al. (2021) was used. After the homogenization process, samples were quickly preserved in cooled bags until delivery to the laboratory. A detailed description of the homogenization process is provided in Supporting Information (SI) (Section 5).

For both water and sediment samples, all equipment in contact with the samples was rinsed thoroughly prior to sampling, using water collected from the stream at the respective sampling sites. Equipment blanks for ISCO autosamplers were carried out with PFAS free water (Supelco–LiChrosolv® - water for chromatography-LC–MS Grade) at two specific temperatures: 4 °C and 25 °C. No PFAS were quantified in the blank samples, except for the equipment blank carried out at 25 °C, in which the concentration of PFBA was 2.4 ng/L, yet PFBA was not quantified in all samples collected during the warmer seasons in water samples. Nevertheless, the results relating to PFBA should be interpreted with caution, taking this variability into account.

2.3. Sample analysis

Water samples were sent ALS Scandinavia AB to be analyzed for 34 targeted PFAS using liquid chromatography-mass spectrometry (LC–MS/MS) based on either US EPA Method 537 and CSN P CEN/TS 15968 or US EPA 533 (Table S10). Water samples were also analyzed for total suspended solids (TSS) according to the standard method SS-EN 872:2005 using vacuum filtration through 1.6 μm glass fiber filters, and zinc (Zn) according to SS-EN ISO 17294-2:2016 and US EPA Method 200.8:1994 after autoclave assisted digestion in 1.2 mL 14 M HNO_3 per 12 mL sample, prior to analysis using inductively coupled plasma-sector field mass spectrometry (ICP-SFMS). Sediment samples were submitted to Eurofins Environment Testing Sweden AB following sample collection and were analyzed using LC–MS/MS for 35 targeted PFAS based on DIN 38414-14 and Powley et al. (2005), and 30 total oxidizable precursors assay (TOP assay) based on Houtz and Sedlak (2012), described in detail in Table S10. All analyses were carried out by laboratories accredited by the Swedish Board for Accreditation and Conformity Assessment (SWEDAC), which follows international QA standards and implements quality assurance/quality control measures.

Analyzed compounds in this study included fourteen PFCAs, ten PFSAs, three Fluorotelomer sulfonates (FTS), three Perfluorinated sulfonamides (FOSA), three Perfluorinated sulfonamidoacetic acids (FOSAA) and two Perfluoroalkyl sulfonamidoethanols (FOSE). Targeted PFAS analysis included the same PFAS for both water and sediment samples, except for one compound, PFHxDA, which was only analyzed in the sediment samples. Table S11 lists the groups, names, abbreviations, CAS Number, formula, chain length and molecular weight, and reporting limit (RL) for the PFAS analyzed in this study. The RL was elevated for some samples due to matrix effects. For compounds with higher RLs, the Table S11 specifies a range of RL values.

2.4. Data analysis

The data were grouped by event type (DW and WW) for water samples and by analysis method (targeted and TOP assay) for sediment samples. First, the descriptive statistics, including percent quantification (p_{quant}), minimum, maximum, and median concentrations were calculated for compounds based on event type. Matrix effects in some samples led to higher reporting limits for certain compounds and, in some instances, these elevated reporting limits exceeded the maximum reported concentrations, resulting in a left-censored dataset. Maximum reported concentration and elevated RL for each compound under DW and WW conditions are presented in SI (Table S12). To address the left-censored data, appropriate statistical methods for left-censored data (Helsel, 2012) were used, employing the Non-detects and Data Analysis for Environmental Data (NADA and NADA2) package in R (Julian and Helsel, 2024).

Concentrations were further categorized by sampling location—upstream (A), directly downstream (B), or further downstream (C)—and by city (Uppsala, Norrköping, and Söderköping), with descriptive statistics calculated for each sampling site. Median concentrations were calculated using the robust regression on order statistics (ROS) method (Helsel, 2012). Differences between sampling sites within the water body for a specific event type were assessed with the non-paired signed -ranked Wilcoxon test. Statistical differences between event types within the same sampling site were assessed with the non-paired Wilcoxon test. All the statistical analyses, including descriptive statistics, were implemented in R, and carried out for compounds quantified in over 20 % of the samples ($p_{quant} \geq 20$).

3. Results and discussion

3.1. Stormwater impact

Along with PFAS, this study also analyzed concentrations of TSS and Total Zinc (Zn_{tot}) which are two pollutants commonly present and transported by stormwater, typically increasing in receiving waters when affected by wet weather discharges (Müller et al., 2020; Pamuru et al., 2022). During WW events, TSS in streams can originate from multiple sources, such as sediment resuspension, agricultural runoff, and erosion, all of which contribute to TSS concentrations in streams. However, these sources are typically triggered by stormwater-induced high flows, suggesting that an increase in TSS during WW indicates stormwater runoff as the source. As summarized in the SI (Section 8), downstream of urban areas, median TSS and Zn_{tot} concentrations were, in general, higher during WW compared to DW. Median Zn_{tot} concentrations ranged between 4 and 51.7 μg/L and exceeded the Predicted No-Effect Concentration (PNEC) for zinc (7.8 μg/L) (NORMAN Database System, n.d.) at most sampling sites during WW events. Upstream of the cities (UppA and NrkA), no increase in these compounds' concentrations was observed during WW. These findings provide evidence that urban stormwater discharges clearly deteriorate the water quality in these urban streams during WW. Further, the expected elevated Zn_{tot} and TSS concentrations during WW also confirm that the samples taken during WW events were indeed capturing the water when impacted by stormwater runoff.

3.2. PFAS concentration and occurrence under dry and wet weather conditions in urban streams

Across all streams, median total quantified PFAS concentrations ranged from 1.42 to 14.91 ng/L in DW samples and from 2.66 to 67.8 ng/L in WW samples, showing an increase in median total PFAS concentrations in WW samples by 1.7–4.5 times compared to DW samples across the sampling sites (Table S15). Notably, the median concentrations of almost all quantified PFAS in WW samples were higher (1.1–5.7 times) than those in DW samples (Table S12).

PFAS concentrations in this study were consistent with other studies carried out in urban streams in Sweden (Nguyen et al., 2022; Sörengård et al., 2022), Singapore (Chen et al., 2017; Nguyen et al., 2011), Japan (Zushi et al., 2008; Zushi and Masunaga, 2009), Switzerland (Müller et al., 2011), and Canada (Meyer et al., 2011). Median concentrations of total PFAS in the heavily urbanized watershed in Singapore were higher (3.7–85.2 times for DW and 2.9–19.7 times for WW) than in this study, with an observed decrease in PFAS concentrations during WW due to already high PFAS concentrations in the DW sample. In contrast, this study found increased PFAS concentrations during WW events, highlighting stormwater as a source transporting PFAS to watercourses. This increase is notable despite sampling in relatively small cities, indicating transport of PFAS by stormwater both from typical urban catchments

and from known sources such as airports. Previous studies carried out in streams passing through urbanized areas, such as Toronto, Canada (Meyer et al., 2011), Yokohama, Japan (Zushi et al., 2008; Zushi and Masunaga, 2009), and areas near Zürich, Switzerland (Müller et al., 2011), reported increased concentrations of PFAS, particularly long-chain PFAS, during runoff events. Although these studies did not report total PFAS concentrations, they suggest, similar to this study, that urban stormwater carries PFAS from urban areas to watercourses.

In the DW water samples ($n = 32$), 13 out of 34 targeted PFAS were quantified in at least one sample (Fig. 1). Of these, the legacy PFAS, PFOS and PFOA were the most frequently quantified compounds ($p_{quant} > 80$ %). Other PFAS with a high quantification frequency ($p_{quant} > 70$ %) in DW samples included two PFCAs (C5-C6: PFHxA and PFHpA) and two PFSAs (C4-C6: PFBS and PFHxS). Out of the 13 quantified PFAS, seven were infrequently quantified ($p_{quant} < 20$ %) in DW samples (Table S12). All frequently quantified PFAS in DW samples, except for the legacy PFAS, PFOS and PFOA, were short-chain PFAS, probably due to their lower hydrophobicity, as indicated by their lower K_{ow} values (Ahrens et al., 2010).

During WW, the variety of PFAS increased, with 17 out of 34 targeted PFAS quantified in WW samples ($n = 67$) (Fig. 1). All PFAS quantified in DW samples were also found in WW samples, notably with higher quantification frequencies. A clear pattern emerged with long-chain PFCAs, such as PFNA, PFDA, and PFUnDA, as well as the precursor 6:2 FTS, being more frequently quantified during WW events (Fig. 1). Additionally, C10 PFCA, C7 PFSA, 8:2 FTS, and EtFOSA, which all are long-chain or particle-associated PFAS, were exclusively quantified in WW samples (Fig. 1). This suggests stormwater likely transports these longer-chain and particle-associated PFAS into aquatic systems, as supported by increased TSS concentrations measured in WW samples in this study. Further, this hypothesis is supported by significant correlations (Kendall's tau test and Spearman $p < 0.01$, rho/tau >0.30, Table S16) between TSS and both long-chain PFCAs (C7–C9: PFOA, PFNA, PFDA) and 6:2 FTS in WW samples. Notably, weaker correlations were observed between TSS and both PFUnDA and PFOS ($0.01 < p < 0.05$, $0.15 <$ rho/tau < 0.30). Significant correlations were also observed between TSS and short-chain PFCAs (C5–C6: PFHxA and PFHpA) in WW samples, indicating that these PFAS might also be associated with particles and stormwater discharges. Furthermore, the quantification of these substances in studies examining stormwater samples further supports their association with stormwater runoff (Codling et al., 2020; Gasperi et al., 2022; Houtz and Sedlak, 2012; Kim and Kannan, 2007; Xiao et al., 2012).

Despite being phased out in the early 2000s (Directive 2006/123/EC, 2006), legacy PFAS, PFOS and PFOA are still among the most frequently quantified in surface waters (Kurwadkar et al., 2022; Podder et al., 2021) and stormwater runoff (Codling et al., 2020; Gasperi et al., 2022; Murakami et al., 2009). This study is no exception, with PFOS and PFOA being the most commonly occurring compounds in both DW ($p_{quant} > 80$ %) and WW ($p_{quant} = 100$ %) samples, highlighting their historical widespread use and resistance to degradation.

3.3. Spatial distribution and PFAS composition in water

Along all studied water bodies, a consistent trend was observed: both the median concentrations of total quantified PFAS and the median concentrations of almost all individual PFAS were higher during WW compared to DW (Fig. 2, Table S17 for individual PFAS concentrations during DW and WW at each sampling site). The highest median concentrations for total quantified PFAS were consistently observed downstream of the urban areas in WW samples (Fig. 2). Although generally higher PFAS concentrations during WW compared to DW were observed for all streams, the concentrations varied due to the unique characteristics of each stream. In general, total quantified PFAS concentrations in water bodies at various sampling sites under DW conditions ranged from 3.3 ng/L to 84.6 ng/L in Norrköping, RL to 16.7 ng/L in Uppsala, and 1.1 ng/L to 7.2 ng/L in Söderköping. Under WW conditions, these concentrations increased and ranged from 3.9 ng/L to 102.3 ng/L in Norrköping, 4.1 ng/L to 33.7 ng/L in Uppsala, and 0.9 ng/L to 10.6 ng/L in Söderköping (Fig. 2 and Table S15).

Across sampling sites, total quantified PFAS concentrations under DW conditions were similar within the same water body, particularly in Söderköping and Uppsala (Fig. 2). PFAS concentrations at the upstream reference site in Uppsala (UppA) were in the same range as the downstream locations along this river, possibly due to discharges from a nearby military air -force base (Nguyen et al., 2022). Further, total median PFAS concentrations directly downstream of the WWTP (UppB2) were approximately 3 ng/L higher than directly upstream (UppB1) for DW, indicating the contribution of WWTP to the concentration of PFAS in the stream. Nevertheless, the PFAS concentrations measured under DW conditions were in the same range along the watercourse. This aligns with previous findings, showing low variability in PFAS concentrations across this stream (Gago-Ferrero et al., 2017). At Norrköping's downstream site NrkC, the total median PFAS concentration during DW was almost three times higher than upstream (NrkA and NrkB), suggesting discharge from potential PFAS point sources between

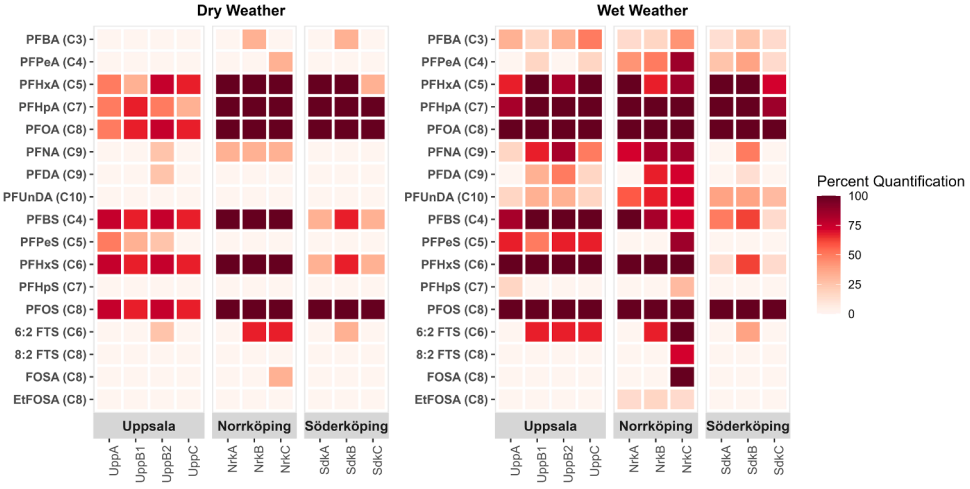

Fig. 1. Percent quantification (p_{quant}) of PFAS under Dry Weather and Wet Weather conditions. Each subfigure corresponds to a stream and is divided into columns representing different sampling sites along the stream. The PFAS are listed on the *y*-axis along with the number of carbon atoms in their structure (in parentheses) and ordered based on chain length from shorter to longer, starting from the top with PFCAs, PFSAs, FTSs, and FOSAs. The percent quantification is represented by the color intensity, with darker shades indicating higher quantification percentages.

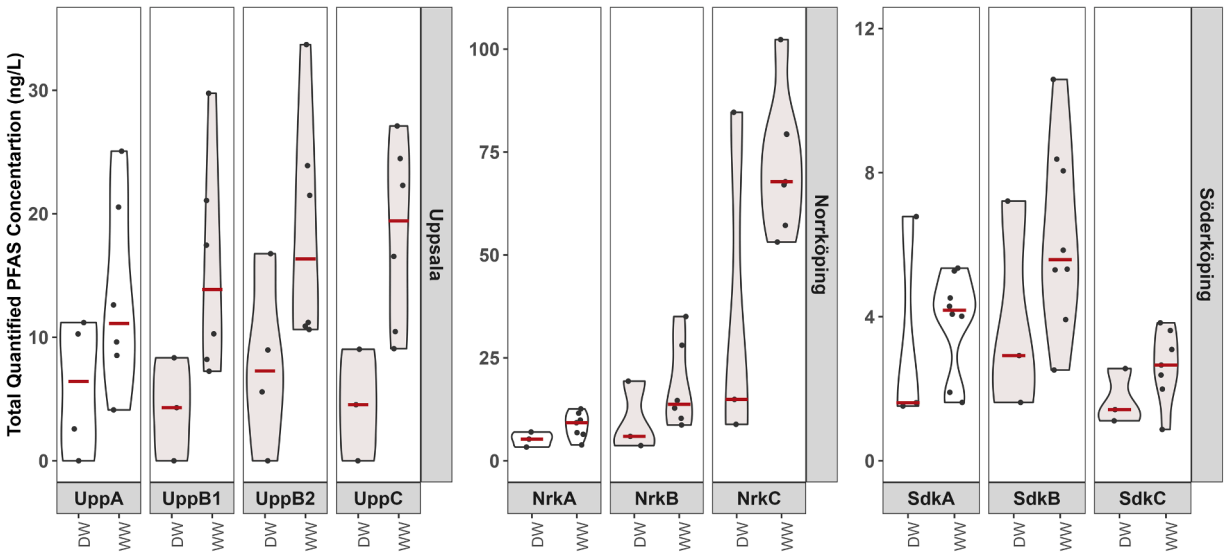

Fig. 2. Minimum, maximum (violin plots), and median (red dash) of total quantified PFAS concentrations (black dots) across streams. Each subplot corresponds to a specific sampling site across the streams, with the *x*-axis differentiating between dry weather (DW) and wet weather (WW) samples, and the *y*-axis representing the total quantified PFAS concentration.

NrkB and NrkC also during DW. Possible sources include the nearby regional airport, an industrial area, and an old landfill site, all located between NrkB and NrkC.

During WW events, an increase of total PFAS concentrations was observed across all sampling sites (Fig. 2) by 1.7 to 4.3 times in Uppsala, 1.7 to 4.5 times in Norrköping, and 1.9 to 2.6 times in Söderköping. The most notable increase between DW and WW was observed at NrkC (67.8 ng/L), where the absolute highest concentration (102.3 ng/L) in this study was measured during one WW event. The total PFAS concentration at NrkC was significantly different from those at NrkA ($p = 0.0017$) and NrkB ($p = 0.0035$) in WW samples, reinforcing the notion of urban stormwater discharges and point sources being drivers for elevated PFAS concentrations during runoff events (Table S18). A significant difference was also detected between the total PFAS concentrations at SdkB and SdkC ($p = 0.011$) under WW conditions. This difference highlights two observations: first, despite the comparably low PFAS concentrations in this stream, the highest levels were detected at SdkB, which is nearest to the urban area, during WW discharges. In contrast to Uppsala and Norrköping, no potential PFAS point source was present here in the catchment, highlighting stormwater as a vector for transporting PFAS from urban areas to streams. Second, a dilution effect seemed to occur at SdkC, the site further downstream, likely due to its distance from the impact of the urban area and another minor stream merging between sampling sites B and C. Interestingly, also at the upstream reference site SkdA, the median PFAS concentration increased 2.6 times to 4.2 ng/L during WW events. This occurred despite its distance from urbanized areas and stormwater outlets, suggesting the influence of potential other unknown sources of PFAS which contribute particularly during runoff events.

At each specific sampling site, differences between total PFAS concentrations under DW and WW were tested. Only at the sampling site located downstream of the urban area in Uppsala (UppC), was a difference of $p = 0.024$ detected, as each WW sample had higher total PFAS concentrations than each DW sample (Table S19). The variability in PFAS concentrations across different WW events led to large variation within each event type, resulting in no statistically significant differences between DW and WW samples at the other sampling sites, although the median PFAS concentrations for WW were always higher than those for DW.

Particularly high total PFAS concentrations were measured in two streams during DW, likely due to a short antecedent dry period. At NrkC, a concentration one order of magnitude higher (84.7ng/L) than other DW samples was observed after two dry days following a 10 mm rain event and 1 mm rain during the sampling. Similarly, two days after 6.7 mm of precipitation and with minimal precipitation (0.6 mm) during DW, in Söderköping total PFAS concentrations at SdkA (6.8 ng/L) and SdkB (7.2 ng/L) were 2.4–4 times higher than in other DW samples. Thus, the observed range of PFAS concentrations was broad at NrkC, SdkA, and SdkB during dry weather (DW) events. This variability led to no significant difference in total PFAS concentrations between DW and WW conditions at these sampling site. Further, it suggests that preceding light rainfall may transport significant PFAS loads to the rivers, with the length of the antecedent dry period impacting PFAS concentrations in streams.

Differences in PFAS concentrations were observed between the three streams, with concentrations consistently highest in Norrköping, followed by Uppsala, and Söderköping, under both weather conditions. The stream in Norrköping showed particularly high variations between the three sampling sites (NrkA, NrkB, NrkC) especially for WW. This water body is not just narrow and shallow, but also partly culverted and relatively small compared to the city, thus behaving more like a storm drain than a stream during runoff events. In contrast, the Uppsala stream maintains a relatively uniform depth and width throughout the study area, despite having a larger catchment and denser urbanized area than that of Norrköping. Meanwhile, in Söderköping, the urbanized area is smaller than the other two cities, yet the river's size is larger than Norrköping and Uppsala. As an estimation of this, the ratio of the total surface area of the river to the total area of the urban catchment was calculated for each river and city and corresponded to 1:20 in Söderköping, compared to 1:74 and 1:80 in Norrköping and Uppsala, respectively. These variations in stream characteristics, alongside other factors such as the extent of urbanization and the proximity to potential point sources of PFAS might be other contributing factors for differing PFAS concentrations observed across sampled streams.

Each watercourse showed a unique PFAS profile, indicating a variability in sources contributing PFAS to receiving waters (Fig. 3). Furthermore, the PFAS composition profiles differed between DW and WW samples, with WW samples showing a more diverse range of PFAS. The common predominant compounds across all streams and both weather conditions included C4–C5 PFCAs (PFHxA and PFHpA), C3–C6 PFSAs (PFBS and PFHxS), and the legacy PFAS, PFOS and PFOA. The predominance of PFOS and PFOA has been reported in many studies of

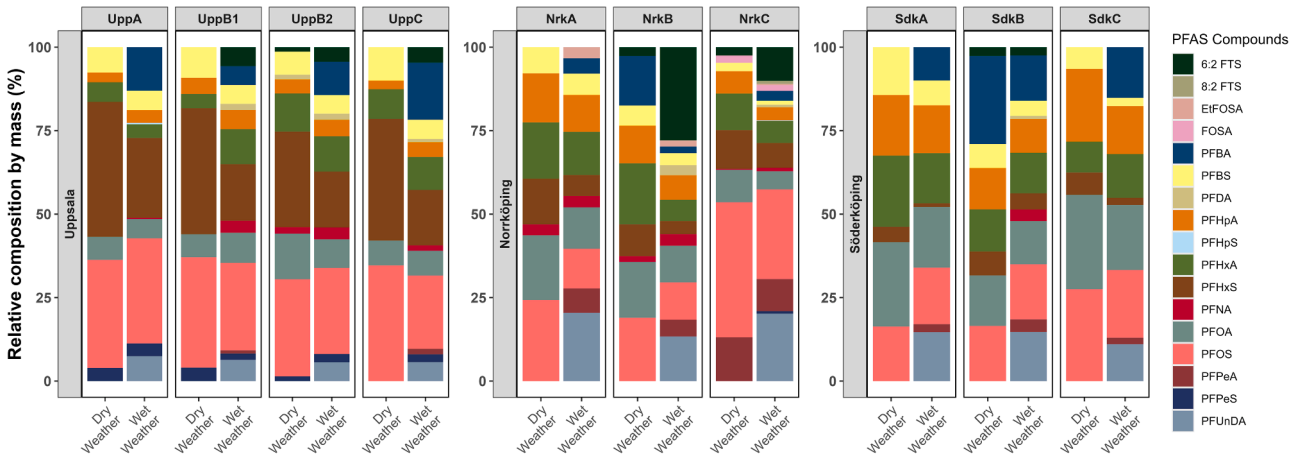

Fig. 3. Relative composition by mass (%) of quantified PFAS across streams. Each subplot corresponds to a specific sampling site across the streams, with the *x*-axis differentiating between dry weather (DW) and wet weather (WW) samples and the *y*-axis indicating the relative composition by mass of the PFAS.

surface water on all continents (Kurwadkar et al., 2022; Podder et al., 2021) highlighting their widespread distribution and persistence in the water phase. Particularly at sites affected by the application of AFFFs (stream in Uppsala and NrkC), PFOS was the most predominant compound, consistent with other studies carried out in streams impacted by such discharges (Ahrens et al., 2015; Anderson et al., 2016; Awad et al., 2011; Chen et al., 2017). The dominance of short-chain PFAS such as PFHxA, PFHpA, and PFBS could be due to the replacement of longer-chain PFAS with short-chain ones in recent years leading to their dominance in the composition profiles (Ateia et al., 2019).

Notably, long-chain PFCA (C10: PFUnDA) was exclusively quantified during WW and was one of the predominant substances in all streams and at all sampling locations. A possible source could be traffic-related, as it has been identified as a major component of automotive lubricant oils (Zhu and Kannan, 2020) and has been found in stormwater from highly trafficked areas in Paris (Gasperi et al., 2022). Similar to the present study, PFUnDA concentrations increased after a rainfall event in Hayabuchi River in Japan (Zushi and Masunaga, 2009). In addition to PFUnDA, compounds such as PFBA, PFNA, 6:2 FTS, 8:2 FTS, EtFOSA, and FOSA altered the PFAS profile in WW, although their dominance was not as pronounced in the overall composition profile.

In contrast to Norrköping and Söderköping, in Uppsala PFHxS emerged as the predominant PFAS. Similar results were reported for the same stream (Nguyen et al., 2022), a water body around Stockholm, Sweden (Ahrens et al., 2015), and several surface water bodies around air force bases in the USA (Anderson et al., 2016). Reasons could be the formulation of AFFF (Anderson et al., 2016) and/or to the long-term exposure of this stream to AFFF and thus the transformation of FTSs to terminal PFSAs (Ruyle et al., 2023). An interesting pattern occurred for 6:2 FTS, known for its frequent quantification in areas affected by AFFF usage (D'Agostino and Mabury, 2017; Ruyle et al., 2021). This compound was not found at UppA during either DW or WW, possibly due to the discontinuation of PFAS-containing AFFF since 2003 at the nearby airport (Gyllenhammar et al., 2015). Also, 6:2 FTS was not quantified under DW (except UppB2), while it was quantified during runoff events at the other three sampling sites, suggesting its ongoing use in urban environments and its transportation through stormwater runoff to the water body.

PFuNDA and PFBA, not quantified under DW conditions, were found across all locations during WW events, indicating their mobilization by runoff. Despite the proximity of the sampling sites UppB1 and UppB2, the PFAS profiles differed under DW sampling. PFNA, PFDA, and 6:2 FTS were not quantified at UppB1 but were present at WWTP-affected UppB2. Moreover, PFOA and PFHxA, two of the most recurrent PFAS in WWTPs worldwide (Lenka et al., 2021), showed increased predominance at UppB2.

During WW in Norrköping, 17 PFAS were quantified at NrkC, which is the highest diversity among all sites, with dominant compounds being PFOS (~27 %), PFUnDA (~20 %), and 6:2 FTS (~10 %). Additionally, PFAS species that were not quantified in other streams in this study, such as 8:2 FTS, EtFOSA, and FOSA, were found in Norrköping, particularly in WW samples.

In Söderköping, the PFAS profiles changed during WW. PFUnDA and PFBA, the latter also quantified in DW samples at SkdB, were only found in WW samples in SdkA and SdkC, comprising a significant portion of the PFAS profile. Additionally, PFPeA was uniquely identified in WW samples in this stream.

Overall, variability in the dominance of PFAS in urban streams indicates local differences in sources and uses of PFAS but tracing the source of each compound is complex and beyond the scope of this work. The occurrence of these compounds in stormwater samples from highly trafficked areas (Gasperi et al., 2022), a landfill site (Chen et al., 2024), street dust (Ahmadireskety et al., 2022), urban street runoff (Murakami et al., 2009), and residential and/or commercial/industrial areas (Codling et al., 2020; Kim and Kannan, 2007; Paige et al., 2024; Xiao et al., 2012) points to their ubiquity and variability in both concentration and occurrence in urban areas. Also, although various relevant sources for these compounds have been identified (including building and construction materials, AFFF, coatings, paints, and personal care products (Glüge et al., 2020; Podder et al., 2021), the exact source of these compounds within urban catchments with many potential known and unknown sources remains unclear, underscoring the need for more research to identify the potential sources of PFAS in urban environments.

3.4. *Site-specific factors*

The correlations between site-specific factors (e.g. precipitation depth, antecedent dry days, flow) and PFAS concentrations were tested across all sampling sites. The SI (Table S20) shows correlations for each sampling site. Each stream had unique relationships with these parameters. Generally, PFAS concentrations (6:2 FTS, PFHxA, PFHpA, PFOA, and total PFAS) were positively correlated ($p < 0.01$ for strong and $0.01 < p < 0.05$ for weak correlation) with precipitation depth at Uppsala sites, except the upstream site (UppA). This suggests that a high level of precipitation, and thus high flows, increase wash-off of accumulated PFAS from urban surfaces. In Söderköping, weak to strong negative correlations were found between PFAS (PFOS, PFOA, PFHxA, PFHpA, PFHxS) and antecedent dry days, indicating longer dry periods are associated with lower PFAS concentrations. Reasons for these negative correlations remain unclear as they are contrary to the expectation that longer dry periods would lead to more accumulation on urban surfaces

and thus higher concentrations of PFAS. No specific pattern of correlation with the site-specific factors was observed for the Norrköping sites.

Interestingly, although precipitation depth and flow rate represent similar variables, strong correlations with PFAS concentrations were observed for precipitation depth. This discrepancy highlights the complexity of factors influencing PFAS dynamics in stormwater and rivers. Given the very large number of variables potentially impacting PFAS concentrations simultaneously, and different behavior of different PFAS, the observed correlations should be interpreted with caution. Despite a rather comprehensive dataset, due to these complexities, explaining such complex factors with the available sample size exceeds the statistical power.

3.5. PFAS in sediment

Of 35 PFAS analyzed, 10 were quantified in the sediment in Norrköping, seven in Uppsala, and only one, PFOS, in Söderköping. Total PFAS concentrations in sediment samples ranged from 0.08 to 4.28 µg/kg, and from 0.8 to 3.2 µg/kg dw, in Uppsala and Norrköping, respectively. In Söderköping, the range was 0.06 to 0.13 µg/kg dw, one order of magnitude lower. Concentrations of each quantified compound are presented in Fig. 4 and Table S21. The total PFAS concentration range in this study was 3.5–26 times lower than in sediment from an urban water body in Singapore (Chen et al., 2017), 4–140 times lower than along the Yellow River in China (Zhao et al., 2016), and 2–4 orders of magnitude less than sediment from Reno/Sparks and Las Vegas metropolitan areas, Nevada, USA (Bai and Son, 2021). The lower PFAS concentrations observed in this study can be attributed to the smaller urbanized areas and size of the connected catchments to the rivers in Norrköping, Uppsala, and Söderköping. This produces less industrial and urban impact on the rivers compared to more heavily urbanized and industrialized areas as in Singapore, China, and the USA, thus implying an impact of urbanization and industrial activities on PFAS levels in rivers.

Overall, the highest concentrations were found at sampling sites with long-existing point sources of PFAS pollution. In Uppsala, this effect is evident at sampling site UppA (3.22 µg/kg dw), with likely sediment transport downstream. Similarly, the highest total PFAS concentrations in the sediment were observed at NrkC (4.28 µg/kg dw) with the most diverse PFAS profile, encompassing ten PFAS. Notably, the highest number of quantified substances at the UppB2 site can be attributed to the impact of WWTP discharge. Despite the presence of stormwater outlets, relatively low PFAS concentrations (range: 0.06–0.13 µg/kg dw) were measured along the Söderköping stream. This is likely because the stream is less affected by urbanization and lacks large known point sources (e.g., WWTP, airport, landfill, industrial area) found in Uppsala and Norrköping, resulting in less PFAS influx from urban areas, as evidenced by water sample analyses.

Despite the variability in PFAS profiles across streams, PFOS emerged as the predominant compound at all sites and was the only quantified compound at the sampling sites without known point sources (e.g. airports), except at NrkA where PFNA was also quantified (Fig. S6). Long-chain PFCA (C13: PFTeDA), along with precursors EtFOSAA and EtFOSE, were not quantified in the water phase but were found in the sediment, although not in all streams.

The TOP assay results presented in Fig. 4 and Table S22 indicate that, at only two sites (NrkC, UppB1), the sum of unknown precursors ($\sum_{\text{unknown-precursors}}$) was higher than $\sum_{\text{targeted}}$, suggesting the presence of many unknown precursors, potentially transforming into PFCAs (C5–C9). In contrast, for five sampling sites (NrkA, NrkB, SdkA, SdkB, and SdkC), $\sum_{\text{unknown-precursors}}$ was below RL. The observed trend at the Norrköping sites (NrkA and NrkB) is likely due to the higher RL of TOP assay compared to targeted PFAS analysis (Table S23). This lower sensitivity could result in underestimating unknown precursors. This phenomenon is observed at the Uppsala sites, where the total of $\sum_{\text{unknown-precursors}}$ is lower than the total of targeted PFAS ($\sum_{\text{targeted}}$).

In general, PFOS is the substance most frequently found in sediment as reported by others (Ahrens et al., 2015; Chen et al., 2017; Nguyen et al., 2016). Notably, long-chain PFCAs (C7–C13), PFHxS, and precursors such as EtFOSAA and EtFOSE, show a higher prevalence in sediments than in water samples. This predominance in sediments can likely be attributed to their hydrophobic characteristics and, thus, sorption to sediments (Ahrens et al., 2010; Nguyen et al., 2016; Zhao et al., 2016). Moreover, PFAS species such as PFOS, PFBA, PFOA and EtFOSE, have also been found in sediments from stormwater ponds (Crane, 2019; Flanagan et al., 2021; Olmsted et al., 2021), gully pots (Wei et al., 2023), biofilters (Beryani et al., 2024) and street sweeping sediment (Ahmadireskety et al., 2022) suggesting their association with stormwater discharges and tendency for accumulation in soil or sediment media. For the TOP assay analysis, although numerous studies

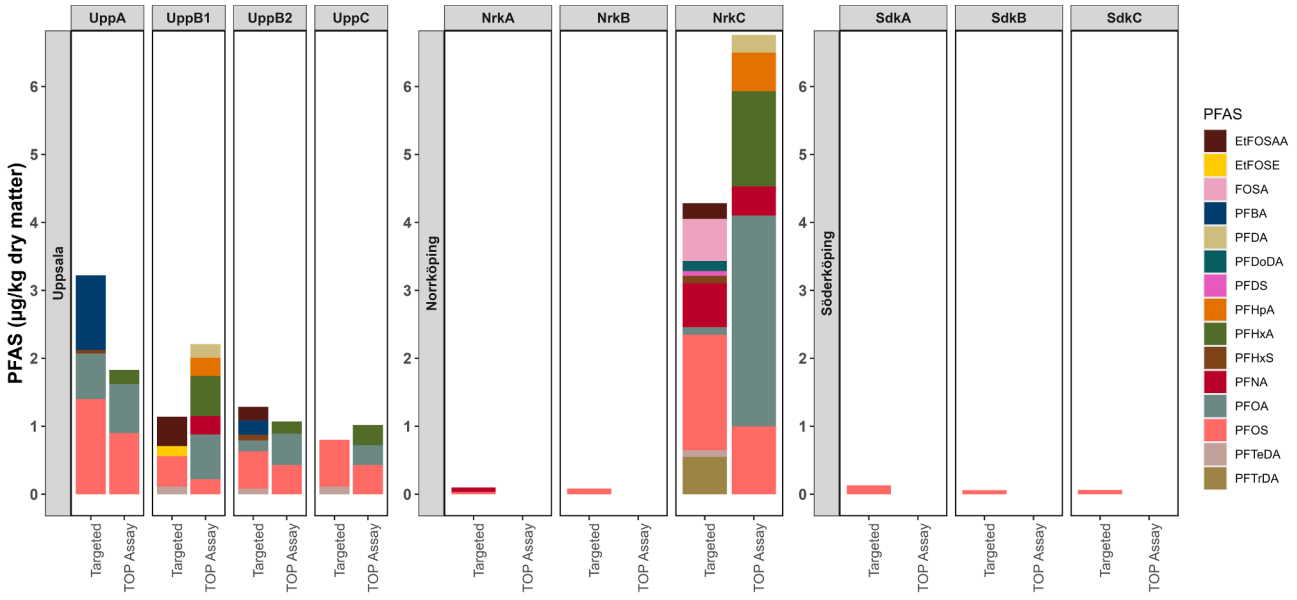

Fig. 4. Concentration of quantified PFAS in sediment across streams. Each subplot corresponds to a specific sampling site across the streams, with the *x*-axis differentiating between targeted PFAS analysis (Targeted) and Total Oxidizable Precursors analysis (TOP Assay), and the *y*-axis indicating the concentrations in µg/kg dry matter.

suggest that sampling sites with elevated PFAS levels often exhibit increased levels of unidentified precursors (Göckener et al., 2023; Houtz and Sedlak, 2012; Macorps et al., 2023), the present study was unable to identify this pattern. This could be due to the higher RL in the TOP assay, which may have hindered the quantification of these precursors.

3.6. Environmental implications

PFOS, which is listed as a persistent organic pollutant and is the only one of the PFAS with a set Annual Average Environmental Quality Standard (AA-EQS) (Directive 2013/39/EU, 2013) for surface waters, was one of the most frequently quantified compounds in this study. Median PFOS concentrations consistently exceeded the AA-EQS (0.65 ng/L) by 2–8 times during DW, and 1.4–29.7 times during WW, except in Söderköping, where the AA-EQS was only exceeded at SdkB during WW (Fig. S7). Also, for the other sites, the degree of exceedance was higher in WW samples. The PFOS prevalence in the rivers is likely attributed to historical uses of PFOS (Podder et al., 2021). Stormwater discharges cause additional concern due to the pronounced exceedance of the AA-EQS during WW in streams, particularly when affected by urbanized areas.

Recently, it has been suggested that the sum of PFAS (sum of PFOA equivalents) be added to the list of priority substances, with an AA-EQS of 4.4 ng/L for surface waters (Directive 2020/2184, 2020). The sum of PFOA equivalents analyzed in this study with the suggested AA-EQS are presented in SI (Section 20). The sum of PFAS was either close to the AA-EQS or exceeded it by 1.1–4 times under DW conditions at all sampling sites, except in Söderköping. Under WW, the proposed AA-EQS was exceeded 2.6–22.9 times in Norrköping and Uppsala. In Söderköping, only SdkB exceeded the AA-EQS by 1.9 times. With the highest risk potency factors (RPF) (Directive 2020/2184, 2020), PFNA (RPF = 10), PFDA (RPF = 7), PFUnDA (RPF = 4), and PFOS (RPF = 2), which became more prominent during runoff events in streams, contributed the most to the sum of PFOA equivalent concentrations. Consequently, AA-EQS exceedance was more prominent in WW samples. This highlights a critical concern: stormwater runoff elevates PFAS levels and introduces a broader variety of PFAS into streams, emphasizing the need for action. Addressing this issue requires more data on PFAS levels in stormwater and their potential sources in urban environments. Furthermore, FTSs (6:2 FTS and 8:2 FTS), quantified in the streams during WW events in this study, are currently unregulated and not listed among the compounds for which AA-EQS have been suggested. Their potential to degrade to terminal PFAS (Ruyle et al., 2023) raises additional concerns, suggesting that developing standards for these compounds in surface water bodies should also be considered.

Given the absence of established AA-EQS for PFAS or their cumulative concentrations in sediments, the observed concentrations of PFAS in sediment samples (targeted and TOP) were compared to the lowest available PNEC (NORMAN Ecotoxicology Database, n.d.) where available, as detailed in SI (Tables S20 and S21). The lowest PNEC value, identified for PFOS at 0.012 μg/kg dw, was exceeded in all streams and sites for both targeted analyses and TOP assay. Other quantified PFAS did not exceed their respective PNEC values. However, it should be noted that the PNEC values for those quantified PFAS were three to five orders of magnitude higher than those of PFOS. This variance in PNEC values might be related to their derivation method, which relies on deterministic methods, thus including uncertainties (Soil and Sediment Risk Assessment of Organic Chemicals, 2004). This inconsistency suggests that current PNEC values may not completely reflect the environmental impact of PFAS in sediment. More research is necessary to improve understanding of PFAS impacts on sediment, thus informing the establishment of AA-EQS values.

4. Conclusions

This study investigated the occurrence, concentration, and spatial distribution of 34 targeted PFAS in three urban streams under dry and wet weather conditions. Findings suggest an impact of stormwater discharges on PFAS concentration across all sampled urban streams. Concentrations and occurrence patterns of PFAS varied between DW and WW, with higher concentrations consistently found under WW, especially at sites downstream from urban areas and point sources such as airports and landfill sites. WWTP contributions to PFAS levels were also evident for both weather conditions, yet not as pronounced as stormwater discharges. Notably, PFOS concentrations exceeded the AA-EQS limit at multiple sites, particularly during WW and in downstream urbanized areas. The sum of PFAS (PFOA equivalents) frequently exceeded the proposed AA-EQS, highlighting stormwater runoff as a major PFAS source to receiving water bodies. Moreover, the findings highlight that monitoring only a few PFAS (i.e. PFOS) is insufficient, given the variety of PFAS detected, their widespread urban use, and transport by stormwater. Interestingly, while short-chain PFAS were more prevalent during DW, WW samples contained more long-chain PFCAs and 6:2 FTS, suggesting stormwater may introduce longer-chain, particle-associated PFAS to the receiving water bodies. The detection of unregulated FTSs like 6:2 and 8:2 FTS in WW samples points to the need for regulatory action, as these compounds can degrade into more persistent PFAS in the environment.

Additionally, river bottom sediment samples were analyzed for 35 targeted PFAS and 30 PFAS with TOP assay analysis. Sediment quality indicates legacy pollution, as sampling sites close to point sources (e.g. airports) show higher concentrations and a greater variety of PFAS. Long chain PFAS (e.g. C7–C13 PFCAs, C6 PFSA) and precursors (e.g. EtFOSAA and EtFOSE) were more prevalent in bottom sediment than in water. Notably, PFOS levels in sediment exceeded the lowest PNEC at multiple sites, indicating potential long-term environmental risks, although current PNECs for other PFAS may underestimate these risks.

The findings show that PFAS are present in the water and sediment phase of the evaluated streams. Apart from point sources, the results clearly indicate that even smaller urban areas contribute to PFAS contamination in urban streams through stormwater runoff. Further research is necessary to understand the sources of PFAS in urban environments and how these compounds interact with stormwater control facilities for protecting surface water quality from potential impacts of stormwater discharges.

Declaration of generative AI and AI-assisted technologies in the writing process

During the preparation of this work, the authors used Grammarly and Microsoft Copilot to check grammar and spelling. After using this tool/service, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

CRediT authorship contribution statement

Suna Ekin Kali: Writing – review & editing, Visualization, Validation, Software, Project administration, Methodology, Investigation, Formal analysis, Conceptualization. **Heléne Österlund:** Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization. **Maria Viklander:** Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization. **Godecke-Tobias Blecken:** Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2024.122973.

Data availability

The dataset supporting this study is referenced at: https://doi.org/10.5878/hpqb-k059.

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