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Per- and Polyfluoroalkyl Substances (PFAS) in Urbanized Section of the Delaware River Watershed: Risk Assessment and Geographical Distribution

Elham Akbari · Tanishka Shah · Kelsey Nazaruk · Rominder Suri · Jeremy Conkle · Gangadhar Andaluri

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Abstract This study investigates the prevalence and risk assessment of per- and polyfluoroalkyl substances (PFAS) in the Delaware River, analyzing 23 water samples collected in 2019 and 2021. The concentration of prevalent chemicals (PFTeDA, PFTrDA, and PFDS) were significantly reduced from average values of 461.67 ng/L, 447.63 ng/L, and 137.10 ng/L between 2019 and 2021, as determined by the analysis of PFAS levels. The most prevalent chemicals in 2021 were PFOA and 6:2FTS, with average concentrations of 5.37 ng/L and 4.23 ng/L, respectively. Based on EPA guidelines, the study assessed environmental and human health hazards from the compounds in the source of drinking water samples using the risk quotient (RQ) and Hazard Index (HI). Following 2016 EPA guidelines, 75% of 2019 and 2021 source water samples had medium risk levels for combined PFOA and PFOS, while the rest were low risk. The RQ of the samples based on 2022 EPA guidelines showed high risk in 92.3% and 38.4% of 2019 collected samples for PFOA and PFOS, respectively. Based on their RQs, all the source water samples in 2021 showed high-risk levels of PFOA. Additionally, the 2023 EPA Hazard Index (HI) approach showed that PFBS, PFHxS, PFNA, and HFPO-DA do not exceed the threshold value. These results underscore the necessity of continuous monitoring and regulation to reduce the adverse effects of PFAS contamination on the Delaware River ecosystem and public health.

Keywords PFAS · Risk assessment · Hazard index · Delaware River

1 Introduction

Per- and poly-fluoroalkyl substances (PFAS) are regarded as emerging compounds posing a significant biological hazard (Calvert et al., 2022). This category of chemicals has been detected in human blood serum (Kotlarz et al., 2020), soil (Brusseau et al., 2020), and water (Phong Vo et al., 2020). Since the 1940s, PFAS have been employed in the production of nonstick cookware, water-repellent textiles, firefighting foam, and water/oil-resistant packaging (Seltenrich, 2020). These compounds can be classified into two groups: long-chain PFAS, which include those with 8 or more carbon atoms for carboxylic PFAS and 6 or more for sulfonic PFAS, and short-chain PFAS, which have fewer carbon atoms in their structure (Pinkard et al., 2024). Short-chain PFAS exhibiting half-lives of approximately 1 to 2 years in blood serum (Nicole, 2020a) and long-chain PFAS

E. Akbari · T. Shah · K. Nazaruk · R. Suri · G. Andaluri (✉)
Department of Civil and Environmental Engineering, Temple University, Philadelphia, PA 19122, USA
e-mail: gangadhar@temple.edu

J. Conkle
Delaware River Basin Commission, Trenton, NJ 08628, USA

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exhibiting half-lives of nearly 3 years in blood serum and up to 94 years in the environment (Mokra, 2021) and have been linked to increased rates of endocrine disruption (Mokra, 2021), cancers (Messmer et al., 2022), liver damage (Costello et al., 2021), infertility (Zhan et al., 2023), birth defects (Stein et al., 2014), thyroid disease (Andersson et al., 2019), and miscarriage (Nicole, 2020b). Considering the long-lasting nature and harmful impact of long-chain PFAS on biological and ecological systems, there is increasing worry about their existence in worldwide water bodies.

Within the Delaware River basin (DRB) there is a growing body of data on a wide range of PFAS compounds (Dunn et al., 2023; MacGillivray, 2021; McCord et al., 2020). PFAS have been quantified in fish tissues, and Delaware Bay bottlenose dolphin plasma (MacGillivray, 2021), as well as the sediment, surface water, groundwater, and vegetation of southwest New Jersey (Goodrow et al., 2020). Targeted and non-targeted studies found that the most prevalent PFAS in the waters surrounding Philadelphia are PFOS, PFOA, PFNA, and PFHxS (Houde et al., 2005; MacGillivray, 2021).

An increasing worldwide effort is being made to enforce more stringent protocols regarding the concentrations of PFAS in drinking water. This regulation strategy is specifically developed to effectively manage and minimize the possible health risks linked to human exposure to PFAS (Liu et al., 2021). In 2016, the United States Environmental Protection Agency (USEPA) issued an advisory establishing a lifetime Health Advisory (HA) threshold of 70 ng/L for PFOA and PFOS in drinking water (USEPA, 2016a). In 2022, USEPA proposed lower interim HA levels at 0.004 and 0.02 ng/L for PFOA and PFOS. In 2021 GenX and PFBS received HA levels at 10 and 2,000 ng/L (EPA, 2022). The USEPA issued final recommended criteria for PFOA and PFOS in 2024 to protect aquatic life in freshwater. The criteria established a Criterion Maximum Concentration (CMC) of 3.1 mg/L for PFOA and 0.071 mg/L for PFOS, with the additional requirement that these limits not be exceeded more than once in a three-year period, on average (USEPA, 2024a). However, there is significant variation observed among individual states, where recommended values for PFOA or PFOS range from 13 to 1000 ng/L (Kurwadkar et al., 2022). Table 1 shows the health-based guidelines for PFAS

Table 1 Guideline concentrations of PFAS in drinking water from various Countries/States

country	Date and State	PFOS (ng/L)	PFOA (ng/L)	PFHxA (ng/L)	PFHxS (ng/L)	PFNA (ng/L)	PFBS (ng/L)	HFPO-DA (ng/L)	Ref
USA	EPA (2022)	4.0	4.0		Hazard Index 1.0 (Unitless)	Hazard Index 1.0 (Unitless)	Hazard Index 1.0 (Unitless)	Hazard Index 1.0 (Unitless)	(USEPA, 2022b)
	EPA (2016)	Combined Concentration of 70							(USEPA, 2022b)
	New Jersey (2016)	13	14		13				(Hu et al., 2016)
	Michigan (2019)	16	8	400,000	51	6	420	370	(MPART, 2019)
	California (2019)	40	10			20	5000		(DDW, 2020)
Canada	(2019)	600	200	200	600	20	15,000		(DWSV, 2023)
China	(2023)	40	80						(Wang et al., 2023)

compounds with a proposed advisory level. Furthermore, in 2023, the United States Environmental Protection Agency developed a Hazard Index (HI) assess the potential health risks to humans associated with exposure to a combination of PFAS chemicals. This HI includes PFNA, PFBS, PFHxS, and HFPO-DA, also known as GenX. Additionally, the USEPA revised its Maximum Contaminant Levels (MCL) of 4 ng/L for PFOA and PFOS (USEPA, 2024d). Although there is an adequate amount of data available for assessing the risks associated with PFOA, PFOS, and some other PFAS stated above, the majority of PFAS observed in drinking water require more data to accurately characterize their associated risks (Cordner et al., 2019a).

The primary aims of this study are: (1) to conduct a thorough analysis of PFAS presence in the Delaware River by employing targeted and non-targeted analysis of samples; (2) to assess the potential risk of human exposure to PFAS through drinking water by calculating the cumulative concentrations of PFAS along the Delaware River; and (3) to evaluate the potential health risks to humans resulting from PFAS contamination in river water by comparing the observed levels with established guidance thresholds.

2 Experimental

2.1 Chemicals and Reagents

PFAS standards and mass-labeled internal standards with purity of higher than 98% for the targeted compound were obtained from Wellington laboratories, Guelph, Canada. A detailed description of the chemicals and their abbreviations are available in Supplementary Information (Table S1). Oasis® WAX SPE cartridges were purchased from Waters. HPLC grade Acetonitrile (> 99.9%); methanol (> 99.9%); Sodium chloride (NaCl > 99%); Sodium bicarbonate ($NaHCO_3$ > 99.7%) and HPLC water were acquired from Sigma Aldrich, USA.

2.2 Sampling

In this study, a total of fourteen sampling locations were selected in the tidal Delaware River and its tributaries in 2019 (n = 14) and 2021 (n = 5) (Fig. 1). In 2019, there were 5 sites from the river mainstem (Trenton and River Zones 2–5) and 9 sites from tributaries (Assunpink Creek, Neshaminy Creek, Pennypack Creek, Rancocas Creek, Frankford Creek, Schuylkill River, Mantua Creek, Brandywine Creek, and Christina River). River zones were established by the DRBC and are shown in Figure S1. In 2021, all 5 sites were in the Delaware River mainstem (Trenton, Bristol, Philadelphia, Chester, and Pea Patch Island). It is worth noting that Mantua Creek, Delaware River-Zone 5, Delaware River-Zone 4, Chester, and Pea Patch Island are not located within source water protection areas, while all other locations are considered part of source water protection areas (DRBC, 2024). The sampling sites were selected in urban and industrial areas. Water was collected through grab samples from the surface, and bottom water samples were taken from sites in zones 2–5. For PFAS analysis, subsurface water was collected using 2L HDPE (high-density polyethylene) bottles. Following the collection of the samples, they were stored in coolers to ensure that they remained at a temperature of 4 ± 2 ℃ during transportation to the laboratory for analysis.

2.3 Analytical Methods

2.3.1 Targeted Analysis

For the 2019 collected samples, analysis was conducted using methods developed in our laboratory, which are detailed in previous studies (Shende et al., 2019, 2021). However, the 2021collected samples were analyzed using the draft EPA Method 1633 which can analyze 40 PFAS compounds across multiple compound classes since this method was not released for previous set of samples. For the analysis conducted using draft EPA Method 1633, the samples were homogenized, spiked with internal standards, and the pH was adjusted to 6.5 ± 0.5. Preconditioning involved cleaning SPE wax cartridges with 0.3% formic acid and 1% methanolic ammonium hydroxide. Samples were passed through the activated cartridges and sample containers were rinsed twice with reagent water and formic acid/methanol, then dried under vacuum. The cartridges were then dried, eluted, and extracted. Clean collection tubes were placed inside the vacuum manifold, and the sample container was rinsed with 1% methanolic ammonium hydroxide. Subsequently, the extract was placed into a 1 mL

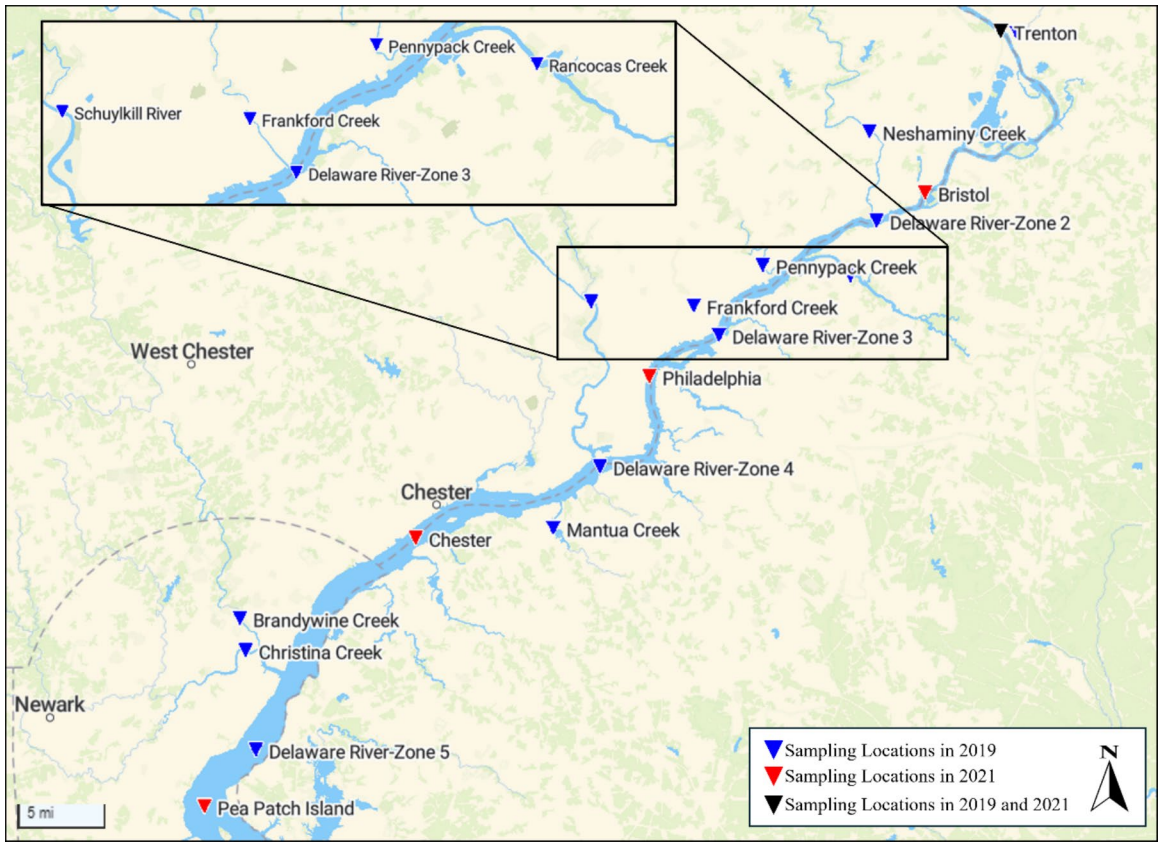

Fig. 1 Sampling sites along the Delaware River in 2019 and 2021

polypropylene vial for analysis using LC/MS/MS (USEPA, 2024b).

2.3.2 *Non-Targeted Analysis*

The 2021 collected samples were subjected to non-target analysis utilizing the SCIEX X500R-QToF system. The samples were analyzed to determine the presence of Solvay replacement compounds. Solvay compounds can serve as alternatives to certain PFAS, such as GenX, and may contain fluorinated elements without being categorized as PFAS. All the samples were suspect screened for chloro-perfluoropolyether carboxylates (ClPFPECA) congeners (DRBC, 2023). Table S2 shows the details of these analytes.

2.3.3 *Quality Assurance and Quality Control (QA/QC)*

An aspect of quality assurance/quality control (QA/QC) was the examination of field reagent blanks (FRB) and laboratory-fortified blanks (LFB), as well as the confirmation of initial precision and recovery (IPR), ongoing precision and recovery (OPR), and continuous calibration check (CCC). Specifically, the QA/QC procedures outlined in Department of Defense (DOD) – Quality systems manual for environmental laboratories have been followed. Table 2 shows the QA/QC criteria (Lordemann et al., 2023; Wingard, 2009).

2.4 Calculation of Risk Quotient and Hazard Index

The risk quotient (RQ) is widely used in environmental risk assessment. The RQ in this study was determined using the following formula:

Table 2 QA/QC Criteria for quantitative analysis of PFAS

QC Check	Minimum Frequency	Acceptance Criteria	Ref
Initial Calibration (ICAL) for all analytes	During instrument setup and after ICV or CCV prior to sample analysis	- RSD for the analyte ≤ 20% - Linear least squares regression for the analyte: $r^2 \geq 0.99$	
Initial Calibration Verification (ICV)	Analyze a second source standard once after each ICAL before doing sample analysis	- All analytes reported fall within confirmed retention time frames - All reported analytes are within a range of ± 15% of the true value	
Continuous Calibration Verification (CCV)	Prior to analysis, at intervals of 10 field samples, and at the completion of the analysis series	- All analytes reported fall within confirmed retention time frames - All reported analytes and substitutes within ± 15% true value	(Lordemann et al., 2023)
Internal Standards (IS)	If utilized, each field sample, standard, and quality control sample	- ICAL Midpoint Standard Requirements: • RT must be within ± 30 s of ICAL midpoint standard • IS signal should be within 50% to + 100% of ICAL midpoint standard	
Method Blank (MB)	One per preparatory batch	- Analyte Detection Limits - • No analytes detected exceeding 1/2 LOQ, 1/10th sample amount, or 1/10th regulatory limit	
Matrix Spike Duplicate (MSD) or Matrix Duplicate (MD)	One per preparatory batch	- RPD ≤ 30%	
Ion Transition Ratio (R)	All samples	- Ion Transition Ratio is calculated from calibration curve confirmation ion peak area to quantitation ion peak area - Ion ratio of the samples should be R ± 3*SD	

$$RQ = MEC/HBG$$

where MEC (measured environmental concentration) is divided by HBG (health-based.

guidelines) for PFAS in drinking water established by various agencies. The four categories for this risk quotient are: very low risk (RQ < 0.01), low risk (0.01 < RQ < 0.1), medium risk (0.1 < RQ < 1), and high risk (RQ > 1).

The Hazard Index (HI) is a useful tool for evaluating health hazards by incorporating the cumulative toxicity of PFNA, GenX Chemicals, PFHxS, and PFBS found in drinking water. The HI is derived as the sum of fractions, with each fraction comparing the measured concentration of a specific PFAS compound in the water to the level at which it is deemed non-hazardous. The HI was determined utilizing the following formula:

$$\text{Hazard Index Value} = \frac{\text{GenX}}{10\text{ ppt}} + \frac{\text{PFBS}}{2000\text{ ppt}} + \frac{\text{PFNA}}{10\text{ ppt}} + \frac{\text{PFHxS}}{9\text{ ppt}}$$

If the running annual average HI exceeds 1.0, the proposed HI Maximum Contaminant Level (MCL) is violated (USEPA, 2024c).

3 Results and Discussion

3.1 Occurrence of PFAS in Delaware River

A comparison of the data from samples in 2019 and 2021 shows the variability in the PFAS concentrations. Table 3 shows the concentration range, median concentration, average concentration, and detection frequency of each PFAS found in all sampling locations during 2019 and 2021. In 2019, the concentrations of PFTeDA, PFTrDA, and PFDS

Table 3 Analysis of PFAS along the Delaware River in 2021 and 2019

Compound	2019				2021			
	Concentration Range (ng/L)	Median Concentration (ng/L)	Average Concentration (ng/L)	Detection Frequency (%)	Concentration Range (ng/L)	Median Concentration (ng/L)	Average Concentration (ng/L)	Detection Frequency (%)
PFPrA	ND[1]	ND	ND	0	ND	ND	ND	0
PFBA	ND	ND	ND	0	0.322–14.74	0.627	3.447	100
PFPeA	0–11.6	0	0.6	5.55	2.079–10.455	3.633	4.699	100
PFMBA	ND	ND	ND	0	0.006–2.471	0.314	0.796	100
HFPO-DA	ND	ND	ND	0	0.035–0.214	0.115	0.113	100
NFDHA	ND	ND	ND	0	ND	ND	ND	0
PFBS	0–8.6	0	1.4	16.66	1.442–6.985	2.233	2.996	100
PFHxA	0–101	0	7.5	5.55	1.047–14.643	1.926	4.365	100
PFPeSA	ND	ND	ND	0	0.02–0.106	0.066	0.0682	100
PFHpA	0–16	2	3.3	72.22	0–11.904	6.46	6.981	80
NaDONA	ND	ND	ND	0	0–0.639	0.307	0.313	80
PFHxS	0–2.2	0	0.5	38.88	0.381–0.924	0.47	0.555	100
PFOA	4–18	5.5	6.4	100	4.07–7.949	6.13	5.802	100
6:2 FTS	ND	ND	ND	0	1.098–9.56	2.9	3.758	100
7:3FTCA	ND	ND	ND	0	ND	ND	ND	0
PFHpSA	0–7.2	0	0.4	5.55	0–0.102	0.045	0.054	80
8:2 FTOH	ND	ND	ND	0	0.025–0.133	0.074	0.077	100
PFNA	0–7.5	0	0.8	27.78	0.384–1.798	1.142	1.027	100
PFOSA	ND	ND	ND	0	ND	ND	ND	0
PFOS	1.1–29	3.4	5.7	100	1.418–5.448	2.098	2.657	100
$PFESA.BP_2$	ND	ND	ND	0	0–0.094	0.027	0.041	60
8-FPDDF	ND	ND	ND	0	0–0.01	0.002	0.003	80
NMEFOSA	ND	ND	ND	0	0–0.13	0.003	0.004	80
PFDA	0–39	0	2.5	16.66	0.256–2.174	0.503	0.808	100
NETFOSA	ND	ND	ND	0	ND	ND	ND	0
PF3ONS	ND	ND	ND	0	0–0.003	0.001	0.001	60
PFNS	0–177	0	13	33.33	0–0.004	0.004	0.004	20
PFUNDA	0–221	1.2	18.9	55.55	0.083–0.346	0.142	0.189	100
NMeFOSAA	ND	ND	ND	0	0–0.006	0.003	0.003	80
PFDS	0–1403	6.3	137.1	72.22	0–0.006	0.006	0.006	20
PFDDA	0–1704	0	189.1	33.33	0–0.076	0.015	0.029	80
$_{11}$Cl-PF3OUdS	ND	ND	ND	0	0–0.002	0.002	0.002	20
PFRrDA	0–2186	240	447.6	72.22	0.00004–0.003	0.00006	0.001	60
PFTeDA	0–1435	395.4	461.7	77.77	0.264–0.663	0.414	0.443	100
PFHxDA	ND	ND	ND	0	0.293–17.684	0.384	3.849	100
PFODA	ND	ND	ND	0	2.863–13.962	3.339	5.950	100

1.Not Detected

were notably high at each sampling site. The average concentrations were recorded as 461.67 ng/L for PFTeDA, 447.63 ng/L for PFTrDA, and 137.10 ng/L for PFDS. The detection frequencies for these compounds were 77.77%, 72.22%, and 72.22% respectively. Additionally, at sampling sites with lower contamination levels, such as Neshaminy Creek, Frankford Creek, and Delaware River-Zone 5,

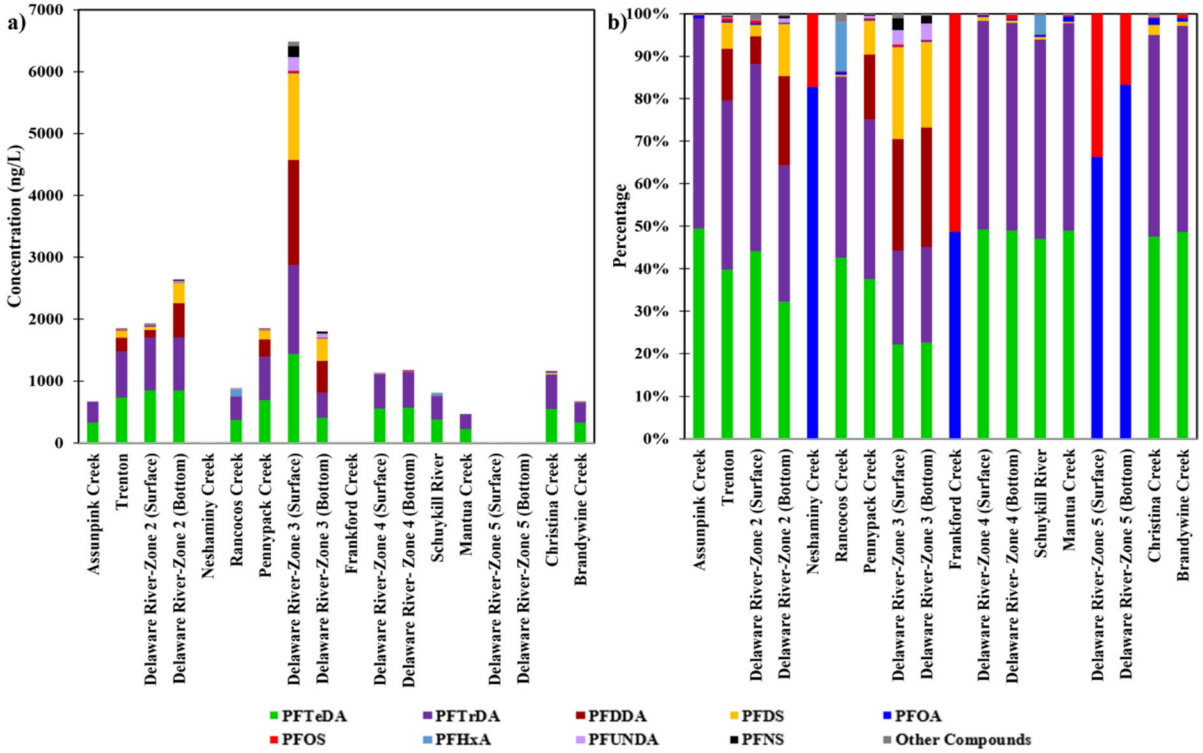

Fig. 2 (a) Total PFAS concentration and (b) Contribution of each individual PFAS in total concentration of PFAS detected at sampling locations in 2019

the total PFAS concentration is predominantly attributed to PFOA and PFOS (Fig. 2). In 2021, 6:2FTS and PFOA had the highest average concentrations of 4.235 and 5.37 ng/L, contributing 10.7% and 12.6% to the total PFAS load, respectively. The persistent substances PFOA and PFOS were detected in all samples collected during both years (Table 3). In 2021, PFODA, PFPeA, PFHxA, PFHxDA, 6:2FTS, and PFBA were detected in 100% of samples, showing a descending average concentration from 5.95 ng/L for PFODA to 3.44 ng/L for PFBA. In 2019, only PFOA and PFOS were detected in 100% of the samples, a variation that could be related to changes in sampling locations, an increase in the number of sampling sites, use of different sampling and analytical methods and temporal differences including varied tidal cycles and river flows (Fig. 3).

In 2019, the maximum ΣPFAS concentration was found in Delaware River-Zone 3 (Surface), followed by Delaware River-Zone 2 (Bottom), which might be a result of the placement of industrial facilities near the different sampling locations (Figure S2). In 2021, the concentration of ΣPFAS in Trenton to Pea Patch Island ranges between 24.107 and 74.492 ng/L. Excluding the Philadelphia sampling site, shows an overall rising trend in Total PFAS concentration as the river flows downstream. The cumulative nature of contamination and the continuous inputs from various sources along the course of rivers tend to result in a possible increase in PFAS concentrations downstream. Rivers accrue water and contaminants from tributaries, groundwater discharge, and surface runoff as they move downstream. Industrial facilities, wastewater treatment plants (WWTPs), and landfills are among the most significant contributors (Marie Amélie Pétré et al., 2021). This pattern was illustrated by a study conducted in Alabama, which showed that the mass flux of PFAS increased consistently as rivers passed through the state. This highlighted the influence of multiple sources and the role of transboundary rivers in the transportation of PFAS from neighboring regions. For example, the Coosa River experienced a 2.2-fold increase in PFAS mass flux downstream, which was due to the inputs from

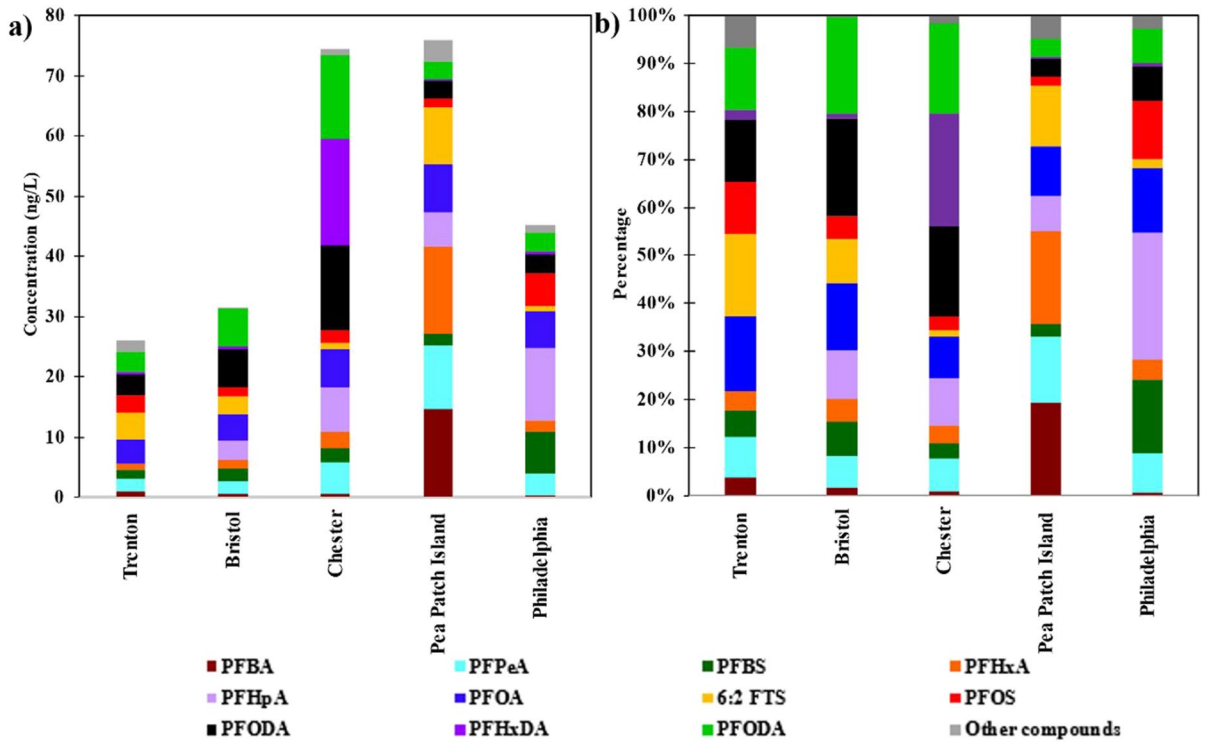

Fig. 3 (a) Total PFAS concentration and (b) Contribution of each individual PFAS in total concentration of PFAS detected at sampling locations in 2021

carpet manufacturing industries, WWTPs, landfills, and military installations. Similarly, the Alabama River experienced a 1.7-fold increase in PFAS mass flux downstream, primarily because of discharges from industries (Viticoski et al., 2022). According to previous studies, the contamination of surface water might be influenced by the proximity of industries to sampling locations near water sources (Crone et al., 2019). Several manufacturers and industries are located near Delaware River that might affect the water quality (Figure S2). For example, water-repellent and stain-resistant fabrics are manufactured by numerous textile mills in the region using PFAS compounds (Viticoski et al., 2022). In addition, PFAS are employed in the production of packaging materials and grease-proof paper (da Silva et al., 2022), with numerous packaging manufacturers located in the vicinity. There are also metal plating and electronics manufacturing facilities in the vicinity of the river, which might be sources of PFAS emissions (Andrews et al., 2021). A 2023 study in Pennsylvania showed that locations with two or more electronics industrial facilities in their vicinity had significantly higher ∑PFAS concentrations in nearby surface waterways (Breitmeyer et al., 2023). Although these sectors may influence PFAS contamination, there is yet no direct data to assess their contributions to the Delaware River.

Moreover, Trenton served as a common sampling location in both 2019 and 2021. Results show a noticeable decrease in PFAS concentrations between these years. Particularly, the concentrations of PFOS decreased from 12.4 ng/L in 2019 to 2.85 ng/L in 2021, while those of PFOA decreased from 6.45 ng/L to 4.07 ng/L. These variations, in addition to fluctuations in total PFAS concentrations, may be related to river flow conditions during the sampling periods. River flow has a substantial impact on the concentrations of PFAS in surface water. In general, higher flows result in dilution and lower PFAS levels, whereas lower flows or drought conditions frequently lead to increased concentrations (Viticoski et al., 2022). Stormwater runoff and precipitation are also significant environmental factors (Tolaymat

et al., 2023). Although the 2019 collected samples were collected at the conclusion of the summer, the 2021 samples were collected in mid-fall, which may have contributed to the observed differences. In addition to hydrological factors, regulatory actions likely contributed to the reduction of PFAS contamination. According to studies, the effective reduction of PFAS levels in the environment can be achieved by phasing out specific PFAS compounds and implementing regulatory measures (Teymoorian et al., 2023). For example, the concentrations of PFOS and PFOA in precipitation samples from the Great Lakes region exhibit an overall decrease between 2006 and 2018, which can be linked to the introduction of regulations and phase-outs during that time (Ehsan et al., 2024). Similarly, the implementation of new regulations and PFAS control guidelines between 2019 and 2021 may have facilitated the effective source control of a variety of PFAS compounds (Crone et al., 2019). Continuous monitoring is necessary to assess the long-term effectiveness of these measures, ensure that PFAS levels are reduced, and monitor the emergence of new PFAS compounds in the environment.

The non targeted analysis showed that in Trenton, PFOA was detected with a high library match of 99.9% but with a low confidence of 34% for PFPrSi. In Bristol, the analysis identified PFBS, 6:2 FTS, and probable hydrocarbon sulfates, all with over 89.6% library match. However, there was low confidence (< 50%) for PFPeA, PFOA, PFPrSi, PFEtS, PFPrS, and PFBA. Chester's results showed the presence of PFEtS, PFPrS, and probable hydrocarbon sulfates, with library matches exceeding 91.3%. The confidence for PFOA and PFPrSi was also low (< 50%). In Philadelphia, 6:2 FTS and probable hydrocarbon sulfates were found with over 75% library match, accompanied by a low confidence (< 50%) for PFPrSi. At Pea Patch Island, the analytes detected included PFEtS, PFBS, 6:2 FTS, PFPrS, and probable hydrocarbon sulfates, with library matches above 84.5% but low confidence (< 50%) for PFPrSi. Additionally, all samples underwent suspect screening for chloroperfluorpolyether carboxylates (ClPFPECA) congeners, however, none were detected in any sample. The Total Ion Chromatogram (TIC) and the mass-to-charge ratios (m/z) for each sampling location are available in the supplementary information (Figure S3 to Figure S12).

The detected concentrations of ∑PFAS in the 2019 collected samples from the Delaware River were among the highest reported in studies of surface waters across the United States. A study conducted in 2021 examined the presence of 17 PFAS compounds in sediment and surface water samples from two local watersheds in Nevada. The total PFAS concentration in the Truckee River water was 441.7 ng/L, whereas the Las Vegas Wash water exhibited substantially higher levels at 2234.3 ng/L. Lake Mead, the downstream reservoir for the Las Vegas Wash, is highly likely to be affected by PFAS contamination from the watershed. Lake Mead is a critical water source for approximately 30 million residents in the southwestern United States, which is why this is particularly alarmingly concerning (Bai & Son, 2021). In the case of the Cape Fear River, a major supplier of drinking water in North Carolina, the average concentration of Σ_{43}PFAS compounds was determined to be 143 ng/L (range: 40–377 ng/L), with an average Σ_{43}PFAS load of 3440 g/day (M. A. Pétré et al., 2022). In addition, the Mississippi River exhibited varying concentrations of PFOA and PFOS at two distinct locations in 2009. The average concentrations of PFOA and PFOS were 17 to 94 ng/L and 15 to 90 ng/L, respectively (Kurwadkar et al., 2022). Trace amounts of PFOA and PFOS have been observed in 58% to 78% of the water systems examined by the New Jersey Department of Environmental Protection (NJDEP) in 2006. A subsequent investigation, which was conducted between 2009 and 2010, analyzed ten PFAS compounds in 31 water systems in this state. The samples utilized in the study consisted of raw groundwater and surface water sources. In general, PFAS were found in 67% of the samples. Among these, PFOA was the most observed chemical, found in about 55% of the samples. 95 percent of surface water samples had PFOA concentrations between 6 and 100 ng/L, whereas 33% of groundwater samples had PFOS amounts between 9 and 57 ng/L (NJDEP, 2014). The Delaware River Basin Commission conducted a study in 2021 and 2022 that specifically examined the water quality of PFAS and assessed the presence of these substances in fish tissue. During the first year of the study, the detection limits for most of the analytes varied between 0.4 and 4.0 ng/L. The data suggested that the majority of the 40 specific PFAS compounds were found to be below the detection limits in this study. PFPeA, PFBS, PHpA, PFHxA, PFNA, PFOA,

PFOS, and PFPeA were identified at Pea Patch Island, out of the six sampling locations along the Delaware River, where the total concentration of these compounds was 40.63 ng/L. This result is consistent with the findings of this study that Pea Patch Island had the highest total PFAS concentration of 75.492 ng/L in 2021 (Conkle, 2023). In the second year of the study, in 2022, at least one target PFAS compound was observed at 14 of the 15 surveyed sites along the Delaware River. The total PFAS concentrations at these sites varied significantly, ranging from 1.9 ng/L to 597 ng/L. The highest concentration was recorded in Bristol, followed by Pea Patch Island, which exhibited a total PFAS concentration of 46.5 ng/L (Conkle, 2024). The PFAS detections and their corresponding concentrations reported in the reports were of a similar value to those observed in the present study. These findings provide essential data on the pervasive occurrence and persistence of PFAS in the environment and highlight the need for continued monitoring and regulation of these substances.

3.2 Risk Assessment

The present work estimated risk quotients (RQ) to improve the understanding of the potential impact of PFAS along the Delaware River. The RQ model has several limitations, including oversimplifying exposure and effects, which can lead to inaccurate risk assessments. It is also inadequate for assessing long-term exposures that may cause significant ecological impacts and lacks quantitative uncertainty, and sensitivity analyses necessary for confident decision-making. Nevertheless, the RQ model provides benefits such as simplicity and accessibility, making it a commonly utilized approach for conducting preliminary chemical risk evaluations. This technique effectively screens chemicals to prioritize them for future evaluation and remains in line with regulatory standards for risk assessments (Karki et al., 2024; Thomaidi et al., 2020). This technique provides a metric to assess the potential risks associated with PFAS contamination in regions used as drinking water sources, covering all sampling site locations except Mantua Creek, Delaware River-Zone 5, and Delaware River-Zone 4 in 2019, as well as Chester and Pea Patch Island in 2021. Based on the guidelines set forth by the EPA in 2016 for combined PFOA and PFOS concentration (USEPA, 2016b), the results indicated that approximately 84.6% of the source water samples analyzed displayed a medium level of risk in 2019, as evidenced by the highest RQ of 0.557 for combined PFOA and PFOS at Delaware River-Zone 3 (Surface). The remaining samples were determined to be within the low-risk range. Furthermore, the risk assessment conducted for the 2021collected samples revealed that only one of the three source water samples showed a medium risk with a RQ of 0.165 in Philadelphia, while the others fell within the low-risk profile. Figure 4 presents the RQs for the 2019 and 2021 collected samples based on the regulations from 2016.

However, there has been a significant emergence of more stringent regulations pertaining to PFAS. In accordance with the regulations established by the EPA in the year 2022 (USEPA, 2022a), it is observed that the risk quotients for PFOA and PFOS concentrations in 2019 fell within the ranges of 0.9 to 4.5 and 0.3 to 7.25, respectively. Delaware River-Zone 3 (surface) exhibited elevated risk levels, followed by Delaware River-Zone 2 (surface) and Trenton, with both PFOS and PFOA RQs classified as high-risk category. Furthermore, 92.3% and 38.4% of the source water samples collected in 2019 exhibited a high level of risk for PFOA and PFOS, respectively (Fig. 5). In 2021, the calculated RQs for PFOA and PFOS concentrations were found to range between 1.02 to 1.5 and 0.35 to 1.4, respectively. This finding presents a significant cause for concern regarding the prevalence of PFOA contamination in the investigated area. Moreover, in 2019, the water samples obtained from more than 15% of the examined sites along the Delaware River exceeded the advisory thresholds established by the New Jersey Department of Environmental Protection for PFOA and PFOS (NJDEP, 2018). Figure 5 presents the calculated risk quotients for all 2019 and 2021 samples, determined using EPA's 2022 guidelines. It is important to note that the health-based guideline applies to drinking water, and source waters will receive treatment prior to use as drinking water.

Based on the Hazard Index (HI) methodology outlined by EPA in 2023 (USEPA, 2024c), and considering the existing data collected in this study, it is apparent that the concentrations of PFBS, PFHxS, PFNA, and HFPO-DA in all sampled areas do not exceed the threshold value of 1.0 as prescribed by the EPA. Additionally, the cumulative sum of these

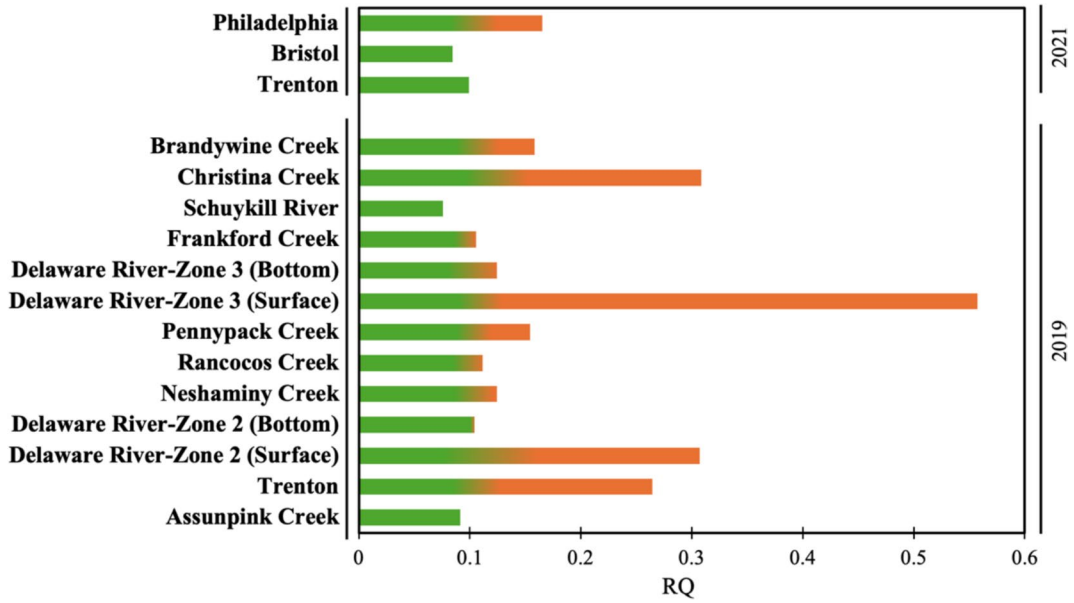

Fig. 4 Risk Quotients (RQ) based on EPA 2016 guidelines for source water samples in 2021 and 2019

substances across all sampling locations is determined to be below 0.2. These collective findings indicate that the contamination levels of PFBS, PFHxS, PFNA, and HFPO-DA present a negligible risk, falling safely within the classification of low risk.

These observations illustrate the growing concern associated with increased levels of these contaminants in the water sources of the region highlighting the significance of evaluating the potential hazards associated with the source of drinking water. In 2024, the USEPA's proposed aquatic life criterion documents conducted a thorough evaluation of PFOA and PFOS toxicity data and established criteria for PFOA and PFOS to safeguard aquatic life from the harmful effects of these substances. The documents also conducted a critical examination of the toxicity data for eight other PFAS, including PFBA, PFHxA, PFNA, PFDA, PFBS, PFHxS, 8:2 FTUCA, and 7:3 FTCA in freshwater environments (USEPA, 2024a). Based on PFAS occurrence data from 2019 and 2021 in the Delaware River, none of the samples exceeded the recommended benchmark values for freshwater. Examining PFAS levels in the Delaware River, which is the source of drinking water for nearby populations, offers essential insights into possible hazards to human health. The findings of this study emphasize the importance of continuous monitoring and regulatory actions to effectively mitigate the potentially harmful effects of PFAS contamination on both the ecosystem of the Delaware River and public health. Therefore, it is imperative to promptly implement control and treatment strategies to minimize the adverse effects caused by these substances.

3.3 Summary of PFAS Health Effects

While drinking water treatment can effectively remove certain PFAS, there remains a risk of these compounds entering drinking water from source waters (Chow et al., 2021). Advanced treatment technologies, including activated carbon, anion exchange resins, and high-pressure membrane systems, have shown efficacy in eliminating PFAS, however, conventional treatment approaches are generally ineffective in reducing PFAS contamination (Crone et al., 2019). These concentrations of PFAS can accumulate in the human body, particularly in blood serum. The significance of risk assessment stems from its role in determining guideline levels that are based on health considerations. During this process, researchers undertake a thorough examination of toxicological, epidemiological, and mode of operation studies to determine the key effect, which relates to the adverse endpoint that is most vulnerable and relevant

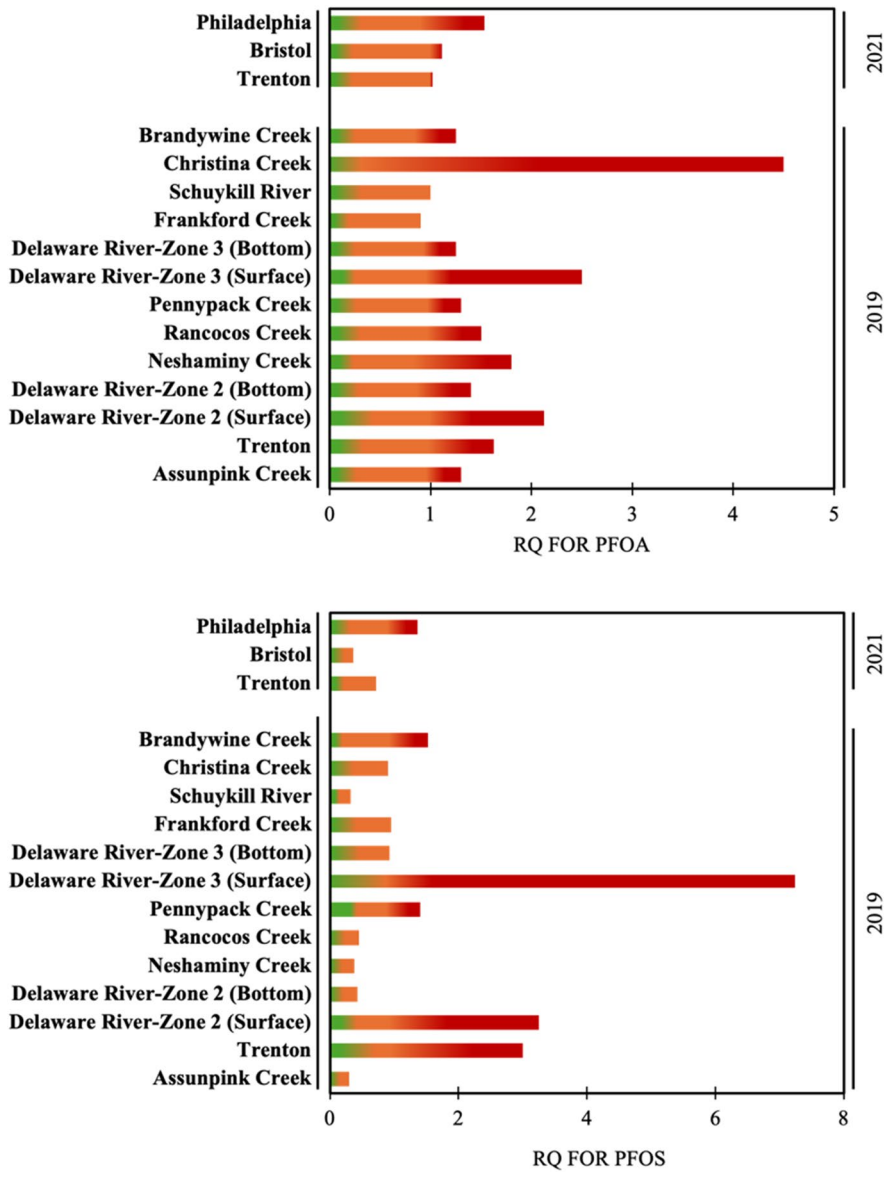

Fig. 5 Risk Quotients (RQ) based on EPA 2022 guidelines for source water samples in 2021 and 2019

to human health (Cordner et al., 2019b). Owing to their widespread utilization, a significant proportion of individuals residing in the United States exhibit measurable concentrations of PFAS substances within their physiological systems (Anderko & Pennea, 2020). The connection between the presence of PFAS chemicals and a variety of health issues has been established, encompassing diseases such as thyroid (Dharpure et al., 2023), kidney (Blake et al., 2018), and liver problems (Attanasio, 2019), as well as elevated levels of cholesterol (Beale et al., 2022). In a case–control study, the serum concentrations of PFAS in newborns diagnosed with congenital hypothyroidism, a rare condition, were compared to those in healthy newborns. In infants diagnosed with congenital hypothyroidism, serum concentrations of several PFAS, such as PFOA, PFNA, PFDA, and PFUnDA, were significantly elevated. Particularly, the concentrations of PFOA were 2.5 times higher in diagnosed newborns (5.40 ng/mL) than in healthy newborns (2.12 ng/mL). The PFNA concentrations in the diagnosed group were 1.93 ng/mL, which was more than three times higher than those in the healthy group (0.63 ng/mL). In addition, the investigation

identified a correlation between thyroid autoantibodies and the concentrations of certain PFAS, such as PFOA and PFHxS. These findings indicate a potential correlation between congenital hypothyroidism and PFAS exposure, which would necessitate additional research (Kim et al., 2016). Another study discovered a correlation between the total cholesterol levels of children and serum PFOA. A 4.6 mg/dL increase in total cholesterol was observed in response to PFOA increases from the lowest to highest quintiles. The study further found that an 8.5 mg/dL increase in total cholesterol was associated with increases in serum PFOS from the lowest to highest quintiles. In young people, the reference level for total cholesterol is less than 170 mg/dL (Sunderland et al., 2018). These studies emphasize the necessity of regulating the levels of PFAS in drinking water to prevent their bioaccumulation in humans and other living organisms. Current regulatory frameworks may be inadequate for addressing PFAS-related issues, predominantly due to the wide variety of compounds that fall under the category of PFAS. In addition, little is known about the hazards associated with low-dose consumption of humans and environmental organisms to these compounds. Enhancing the PFAS environmental assessment requires the development of more sensitive endpoints (Beale et al., 2022). Also, it is crucial to point out that the data provided in this report specifically pertains to surface water, which will undergo additional treatment prior to its consumption as drinking water.

4 Conclusion

This study presents new data about the concentrations of PFAS in the Delaware River. It is crucial to clarify the characteristics and behavior of these substances, specifically regarding their occurrence and risk assessment. The data highlights the greater extent of PFAS contamination in the river, emphasizing that the focus has previously been predominantly on specific PFAS such as PFOA and PFOS. The data indicate a substantial level of risk associated with PFOA and PFOS concentrations in water samples, as per the 2022 EPA guidelines. A large proportion of the samples (92.3%) exhibited high-risk levels of PFOA in 2019, while 38.4% did so for PFOS. As demonstrated by their risk quotients (RQs), all tested samples exhibited elevated levels of risk for PFOA by 2022 regulations. This study reveals that a diverse range of PFAS types have been detected in significant concentrations within the river. The results also reveal the deficiencies in the existing regulatory frameworks, which face challenges in effectively addressing the risks associated with the wide range of compounds categorized as PFAS. The findings of this study highlight the necessity for developing best management practices to prevent PFAS contamination in water bodies. There is a critical need to implement a comprehensive strategy to address PFAS pollution specifically in the Delaware River. Moreover, consistent monitoring of PFAS levels in ambient water is essential to protect human health and the environment from the potential hazards associated with PFAS exposure.

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Data Availability The data that support the findings of this study are available in supplementary information (Table S3 and Table S4).

Declarations

Competing Interest The authors declare that they have no competing interests, including but not limited to personal financial interests, funding sources, employment affiliations, or any other relationships that may inappropriately influence the integrity of work reported in this paper.

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