Science of the Total Environment 878 (2023) 163123

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Review

Environmental and health impacts of PFAS: Sources, distribution and sustainable management in North Carolina (USA)

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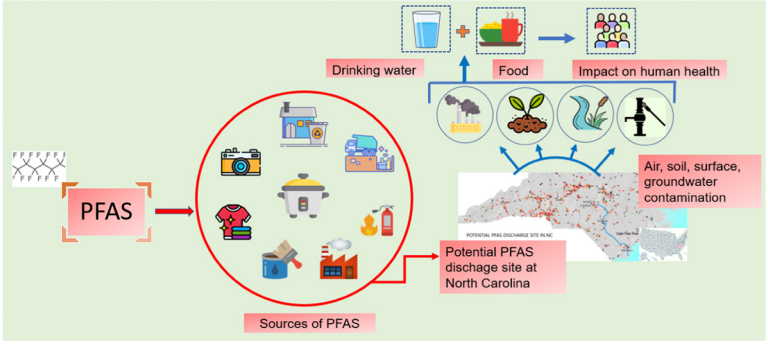
HIGHLIGHTS

- PFAS are widespread in many environmental segments of North Carolina, USA.
- The primary sources of PFAS in North Carolina are identified.
- Drinking water, surface and groundwater, air, sediment, aquatic species, and human blood serum are contaminated by PFAS.
- The adverse effects of PFAS on human health have been reported in North Carolina.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Damia Barcelo

Keywords:
Contamination
Distribution
Environment
Health impact
North Carolina
PFAS sources

ABSTRACT

Poly- and perfluoroalkyl substances (PFAS) are a class of manufactured chemicals that have recently attracted a great deal of attention from environmental regulators and the general public because of their high prevalence, resistance to degradation, and potential toxicity. This review summarizes the current state of PFAS and its effects on the environment of North Carolina, USA. Specific emphasis has been placed to identify i) the sources of PFAS in North Carolina ii) distribution of PFAS in different environmental segments of North Carolina, including surface water, groundwater, air, and sediment iii) drinking water contamination iv) impact of PFAS on human health v) PFAS accumulation in fish and other biota vi) status of PFAS removal from drinking water and finally vi) socioeconomic impact of PFAS uncertainties. Continuous discharges of PFAS occur in the North Carolina environment from direct and indirect sources, including manufacturing sites, firefighting foam, waste disposal and treatment plants, landfill leachate, and industrial emissions. PFAS are widespread in many environmental segments of North Carolina. They are more likely to be detected in surface and groundwater sediments and can enter aquatic bodies through direct discharge and wet and dry deposition of emissions. Eventually, some adverse effects of PFAS have already been reported in North Carolina residents who could have been exposed to the chemicals through contaminated drinking water. Furthermore, PFAS were also found in blood samples from fish and alligators. PFAS were confirmed to be present in water, sediment, organic compounds, and aquatic species at all levels of the food web. However, there is still a substantial amount of work to be done to understand the actual contamination by PFAS in North Carolina comprehensively.

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http://dx.doi.org/10.1016/j.scitotenv.2023.163123
Received 19 February 2023; Received in revised form 22 March 2023; Accepted 23 March 2023
Available online 29 March 2023

Contents

1. Introduction

Per and poly-fluoroalkyl substances (PFAS) are a class of manufactured organofluorine chemicals that are partially or wholly fluorinated surfactants and have been used commercially since the 1940s (Barzen-Hanson et al., 2017; Prevedouros et al., 2006). These compounds have a hydrophobic-lipophilic carbon chain with fluorine atoms in place of hydrogen atoms in some or all of the carbon atoms. In addition, they may be divided into a number of classes and subclasses according to the functional groups at different places, with this classification further incorporated into the long- and short-chain compounds (Ahrens and Bundschuh, 2014). PFAS have widespread use as surfactants, water-repellent sprays, lubricants, additives, paints, and firefighting foams due to the unique capabilities conferred by the electronegative bond in these substances (Adu et al., 2023; Schaider et al., 2017).

Perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) are two of the most widely known and extensively investigated PFAS chemicals. They have been found all over the world in a wide range of environmental segments, including soils, sediments, surface and groundwater, and drinking water (Li et al., 2022; Zhang and Liang, 2022). Both PFAS chemicals have also been linked to a number of negative health effects (Crone et al., 2019; Rahman et al., 2014). Traditional and innovative strong C-F links make PFAS extremely resistant to breakdown by microorganisms, although under suitable conditions (e.g., acidic pH and high concentration of iron), both PFOA and PFOS were demonstrated to be biodegradable (Huang and Jaffé, 2019). The slow biodegradation, if it takes place, results in long half-lives of 8 to 111 years in the soil (Council, 2022). In 2016, in response to concerns about the health implications of these chemicals, the US Environmental Protection Agency (EPA) set a health advisory limit of 70 ng/L for combined PFOA and PFOS in drinking water (Cordner et al., 2019). In June 2022, the interim health advisory level for PFOA and PFOS was lowered to 0.004 and 0.02 ng/L, respectively. These limits are significantly stricter than the 70 ng/L suggested by the EPA for both chemicals in 2016 (Hogue, 2022). They pose risks to human health and the aquifer system because of their relatively high-water solubility and ease of movement to the subsurface environment. It is assumed that the chemicals bioaccumulate and biomagnify as they move up the food chain. Laboratory animals exposed to PFAS in vivo and in vitro investigations demonstrated toxicity in various systems, including reproduction, development, the liver, the nervous system, the immune system, and hormones. Exposure to PFAS occurs when people consume contaminated food or drink, breathe contaminated dust, or transmit the chemical from their hands to their mouths (Grandjean and Clapp, 2015). Obesity, cancer, and suppressed immunity in young people have been linked to harmful chemicals, but very little research has been reported regarding the direct negative effects of PFAS on people (Sunderland et al., 2019).

The characteristics and environmental fate of the PFAS compounds are significantly influenced by the chain length and functional group. Long-chain PFAS and short-chain PFAS are the two categories that are most often used to characterize this class of chemicals. Long-chain PFAS are often referred to as perfluoroalkyl sulfonic acids comprising more than six carbons, such as perfluorooctane sulfonic acid (PFOS), as well as perfluoroalkyl carboxylic acids containing more than seven carbons. The short-chain PFAS contain fewer carbons than their longer-chain counterparts, such as perfluorobutanoic acid (PFBA) (Evich et al., 2022; Zhao et al., 2016). Although shorter-chain PFAS are accumulated more in plants and less likely to bioaccumulate in humans than their longer-chain counterparts, there is a potential that they still present significant risks to both people and ecosystems. Short-chain PFAS seem to be more mobile because they are more water-soluble and have lower sorption to soil particles (Ateia et al., 2019; Bowman, 2015). As a result, they spread further from their original source, affecting the health of individuals in faraway communities. In addition, PFAS with short chains is pervasive in the environment due to their widespread usage and rapid mobility once released (Wang et al., 2015).

The state of North Carolina (NC) is home to >10 million people and serves as a critical habitat for various endangered plant and animal species (Mulrooney et al., 2023). North Carolina's warm and humid environment has helped it become a leading producer of peanuts, sweet potatoes, and other agricultural goods in the United States (Lippmann et al., 2013). If PFAS contaminate these agricultural products, humans, animals, and other visitors to North Carolina may be in danger of PFAS exposure.

Whether this is a possibility or not is unclear at this time due to the lack of detailed knowledge and studies on PFAS' prevalence, origins, fate and movement in the environment.

In view of the limited availability of clean water throughout the world and the well-documented effects of pollution on people and the ecosystem, it is crucial to learn more about the prevalence and toxicity of PFAS in the natural environment. Therefore, the main objective of this study is to identify potential sources of PFAS, the distribution and impact of PFAS in surface water, groundwater, soil and air, and then assess the contamination of drinking water in North Carolina, USA (Fig. 1). It also addresses important knowledge gaps, problems, and the need for more research in the state. Recently published studies on the contamination, distribution, socio-economic impact and environmental effects in North Carolina indicate a growing interest in this field of study. Based on this review, we have made several recommendations on how North Carolina could sustainably manage its PFAS contamination.

2. Hotspot of PFAS: North Carolina

We chose North Carolina for our review because it is one of the most affected states among the 50 in the United States (Barnes, 2020). For example, among the 44 locations examined in 31 states throughout the nation, Brunswick County, North Carolina, has the maximum level of PFAS contaminants in its water supply (EWG, 2020). A large PFAS contamination site was discovered in North Carolina, making it an unfortunate early example in this field (Gecampbe, 2021). The state of North Carolina has been working to become a national leader in addressing the risks associated with PFAS chemicals in drinking water, and environmental regulators have been at the forefront of this effort (Lancaster, 2022). As a result, other states may benefit from looking at the general conditions and consequences of North Carolina PFAS contamination as a forecasting guide.

3. Sources of PFAS in North Carolina

The most common sources of PFAS in the environment can be divided into two types: direct and indirect. The direct sources can be further categorized as follows: industries; metal coating activities; aqueous film-forming foam applications; paper and textile coating, etc. Indirect sources include leachate coming from landfills and effluent from wastewater treatment plants (Dasu et al., 2022). There is strong evidence that large-scale fluoropolymer production facilities are a major source of PFAS in the environment and individuals, especially those who work at these facilities. Therefore, these sites are the main recognized sources of PFAS (Zhou et al., 2022). Additionally, PFAS chemicals have been detected in wastewater treatment plants (WWTP) in North Carolina (Fig. 2), plastic and textile production sites, and fire training grounds where foam extinguishers are used (Table 1) (McCord and Strynar, 2019; Olsen et al., 2017). As a result, higher proportions of industrial locations, military fire training zones, and WWTPs led to higher concentrations of PFAS in water (Hu et al., 2016).

3.1. Manufacturing facilities

Fayetteville Works is home to many different product lines and manufacturing facilities. This manufacturing plant is located on the Cape Fear River's banks, just downstream of Fayetteville, North Carolina (B. Cahoon, 2020). This plant includes the Butacite facility, the Nafion facility, the GenX manufacturing site, and the vinyl ether facilities. This fluorochemical plant previously owned by Dupont, located on 2150 acres in Fayetteville, North Carolina, began to discharge its process wastewater into the Cape Fear River (CFR) in 1980 (Kotlarz et al., 2020) (Fig. 2 A). Since 2015, the Chemours Company has owned the facility. However, Chemours' Director of Product Sustainability stated that GenX in the river is an unexpected and unregulated by-product of industrial facilities. Although emission capture for this chemical is not mandated, abatement technology has been implemented to minimize releases (Chemours, 2017).

3.2. Firefighting foams

PFAS are “forever chemicals” because of their exceptional physical and thermal stability, as well as their chemical persistence. Therefore, they were included in aqueous film-forming foams (AFFFs) for firefighting (Pozo et al., 2022). The constant and recurring use of AFFFs in emergencies

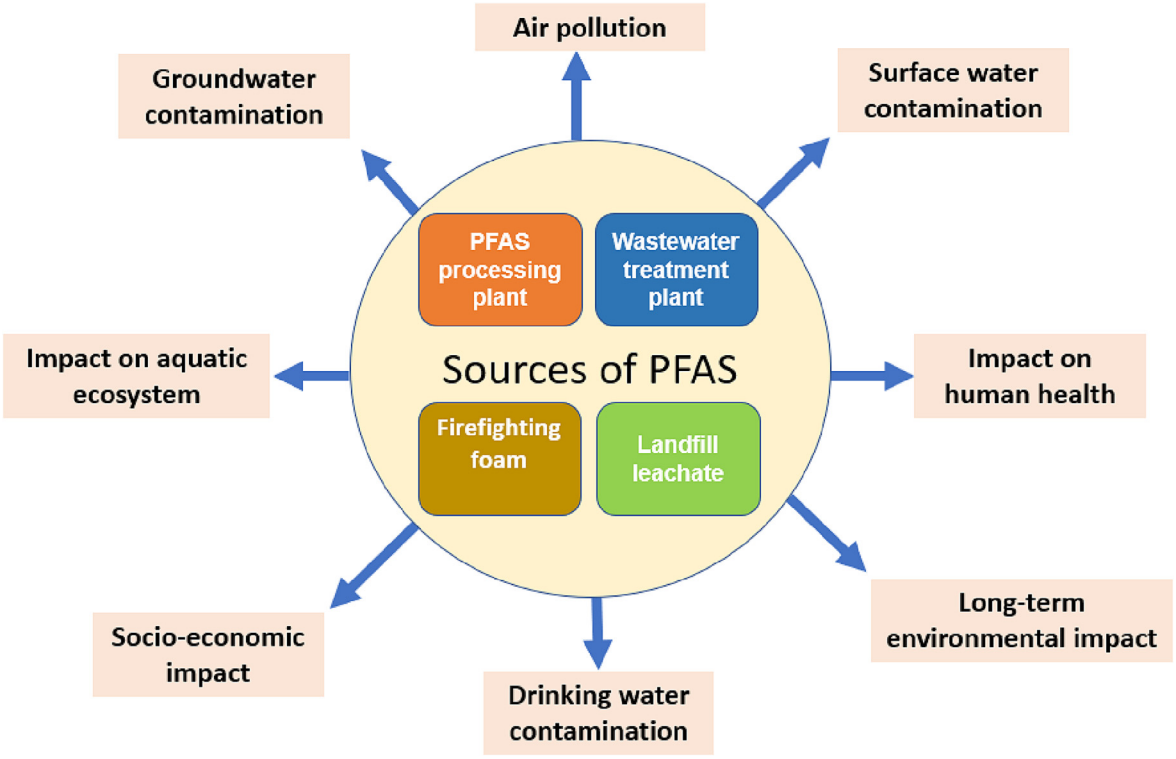

Fig. 1. A diagram describing the sources of PFAS and their impact on human health and the environment.

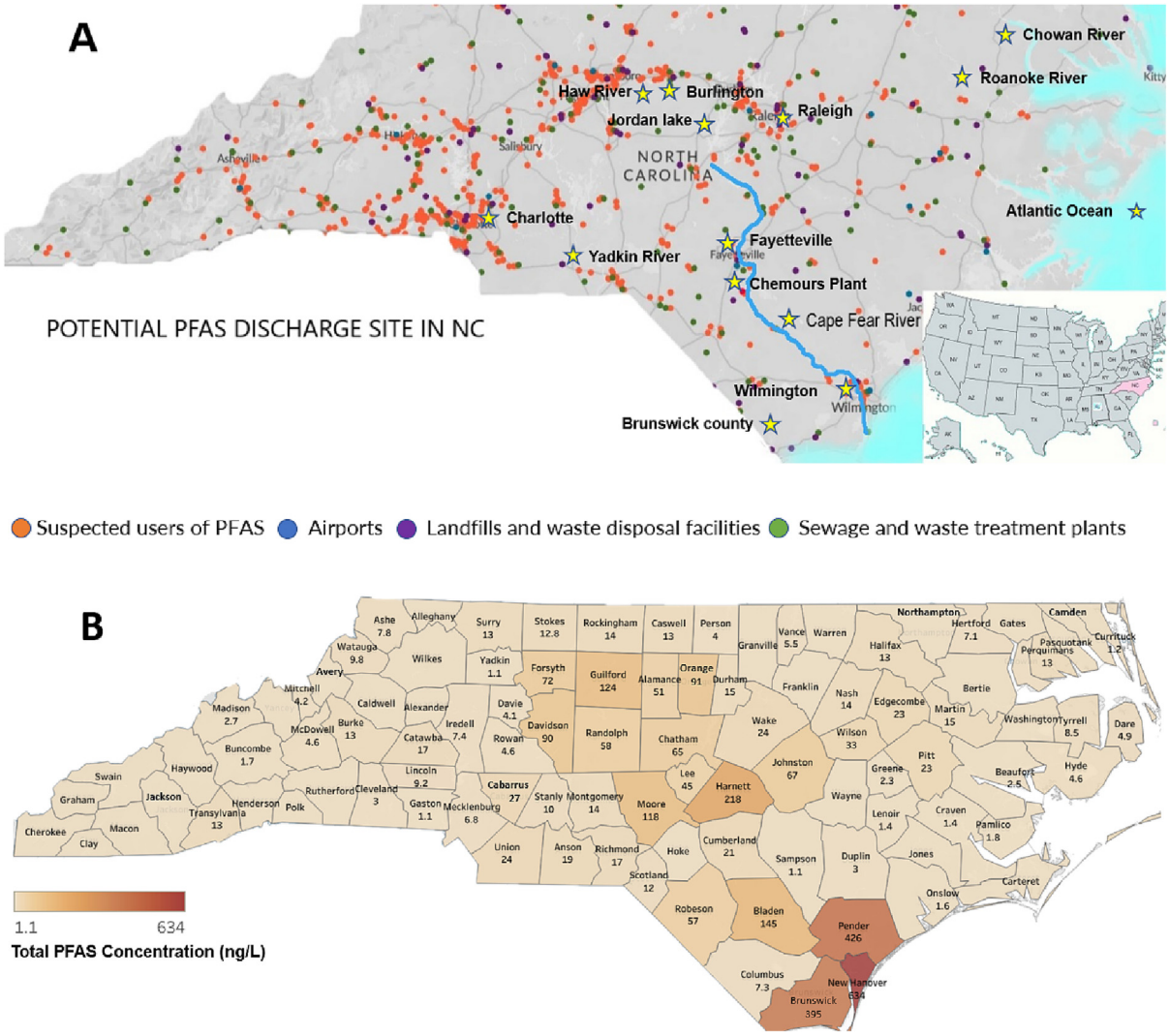

Fig. 2. (A). Suspected locations of PFAS discharge in North Carolina, USA (EWG, 2023a, 2023b). This map shows the areas of municipal and industrial facilities, airports, landfills and disposal facilities, and sewage treatment that have been identified as sources of PFAS. (B) Geographical distribution of PFAS by county in North Carolina's drinking water tested in 2019 measured in (ng/L) (EWG, 2022a, 2022b). The total PFAS mainly includes PFBA, PFBS, PFHxS, PFHxA, PFOS, PFOA, PFPeA, PFHpA.

and for training reasons, especially in military facilities, airports, and municipal fire departments, has led to direct discharges into the environment (Filipovic et al., 2015). The nine PFAS-producing facilities in North Carolina are included in Table 1. Six of these nine sources were found to be potential sources of firefighting foam. These firefighting foams were generated at military bases, fire stations or the state's two airports. For example, in 2017, a PFOS + PFOA concentration of 179,348 ng/L was found in the Marine Corps Base (MCB) groundwater at camp Lejeune in North

Table 1
Location and identified sources of PFAS in North Carolina, USA (EWG, 2022a, 2022b).

Identified sources	Location	Category of source	Detected segment	PFAS	Maximum PFAS level (ng/L)	Year
Charlotte Douglas International Airport	Charlotte	Firefighting foam	Groundwater On-base	PFOS + PFOA	10,100	2019
Stanly County Airport	Stanly County	Firefighting foam	Groundwater On-base	PFOS + PFOA	985	2019
Fort Bragg	Fayetteville	Firefighting foam	Eureka Springs Water System	Total PFAS*	84.1	2016–2019
Private drinking water wells	Fayetteville	Industrial Manufacturing	Private Drinking Water	GenX	4000	2017
The Chemours Company - Fayetteville Works	Fayetteville	Industrial Manufacturing	Groundwater	PFOS + PFOA	3290	2017
Seymour Johnson Air Force Base	Goldsboro	–	Groundwater On-base	PFOS + PFOA	312,000	2017
Camp Lejeune NC, MCB	Jacksonville	Firefighting foam	Groundwater On-base	PFOA + PFOS	179,348	2017–2020
Cherry Point Marine Corps Air Station	Havelock	Firefighting foam	Drinking water Off-base	PFOS + PFOA	151	2017–2018
MCOLF Atlantic	Atlantic	Firefighting foam	Drinking Water On-base	Total PFAS	70	2017

Total PFAS* means sum of the PFAS including PFBS, PFHxS, PFHxA, PFHpA, PFOS and PFOA.

Carolina, which was generated by firefighting foam at that location. Furthermore, 1218 fire departments in North Carolina's 100 counties use or stockpile AFFF (Bodnar, 2021). Although most aircraft crashes occur at or near airports, fires may also happen at gas or oil extraction sites, petroleum plants, storage facilities, chemical manufacturing plants, and railroad crashes, where AFFF would still be anticipated to be used (Salvatore et al., 2022).

3.3. Landfill

Numerous products containing PFAS are disposed of in landfills, and PFAS have been detected in landfill leachate (Lang et al., 2017). Although landfills have been recognized as significant contributors of PFAS to the environment, surface and groundwater around landfills in the United States are typically not monitored for the presence of these chemicals (Walsh, 2020). In one study, 35 samples of surface water near two landfills in Sampson County (SC) and Orange County (OC) in North Carolina were collected and analyzed for PFAS content (Walsh, 2020). Six new (GenX, PMPA, PEPA, NVHOS, Nafion BP2, Nafion BP4) and six legacy (PFBA, PFPeA, PFBS, PFHxA, PFHpA, and PFOA) PFAS were detected in the experiment. These twelve PFAS have concentrations ranging from 33.3 ng/L to 780 ng/L when added together. Legacy PFAS were found in surface water in both landfills, whereas novel PFAS were found only in surface water near the South Carolina dump. The mass of 70 PFAS was tested in 95 samples of leachate from landfills in the United States with various climates and disposal ages. In 2013, between 563 and 638 kg of measured PFAS were estimated to be released into wastewater treatment facilities from US landfill leachate (Lang et al., 2017).

3.4. Household usages

Many common household items contain PFAS and can contribute to PFAS levels in air and dust (Kotthoff et al., 2015). PFAS can be found, for example, in nonstick kitchenware and food packaging (Begley et al., 2005; Schaider et al., 2017; Vandermeyden and Hagerty, 2020), personal care items, such as cosmetics and sunscreens (Harris et al., 2022) and impregnation sprays and compounds used mainly for textile protection, such as stain-resistant upholstery and carpets (Schellenberger et al., 2022). After their useful lives, these PFAS-containing home goods are thrown into the trash and end up in landfills (Liu et al., 2021). Debris from commercial establishments, such as stores, restaurants, and offices (commercial waste) and residential establishments, such as houses, apartments, and condominiums (household waste), is collected by waste collection trucks and sent to a waste treatment facility. Throughout the United States, municipal solid waste landfills receive most of the trash generated from homes and businesses (Renou et al., 2008). However, no data was found on the contribution of household items to PFAS contamination, particularly in North Carolina.

3.5. Waste disposal plants

Since the middle of the 20th century, PFAS, a category of manmade chemicals, have been utilized in a vast array of commercial and industrial applications (Wang et al., 2017). With their desirable properties and manufacturing viability, PFAS are mass-produced for extensive industrial use (Glüge et al., 2020). Consequently, both short- and long-chain perfluoroalkyl acids (PFAAs) were extensively discovered in the influents and effluents of WWTPs, and numerous studies indicated a rise in the amounts of PFAAs after wastewater treatment (Lenka et al., 2021). As a result, PFAS-contaminated water originated from sludge sprayed by Burlington's treatment plant to nearby fields and eventually reached Haw River's streams by runoff (SELC, 2019). The highest concentration of Σ_{13}PFAS (1197 ng/L) tested in the Haw River area had also been found downstream of Burlington, indicating that the PFAS source was likely textile wastewater from the Burlington wastewater treatment facility. The maximum concentrations of PFHxA (perfluorohexanoic acid), PFPeA (perfluoropentanoic acid), PFHpA (perfluoroheptanoic acid), PFBA (perfluorobutanoic acid), PFOA, PFOS were 416.8, 274.1, 235.9, 189.9, 133.3, and 110 ng/L, respectively, in the Haw River area in the year 2019 and 2020. Despite annual changes in overall PFAS content, the effect of release on PFAS levels was reasonably stable (Pétré et al., 2022).

4. PFAS contamination in the environmental segment of North Carolina

The Cape Fear River (CFR) basin (Fig. 2), located in the middle and along the coast of North Carolina, is comprised of approximately 9300 mile^2 of waterways and provides services to about 5.2 million people (Guillette et al., 2022). Several North Carolina rivers, particularly the CFR, have been found to contain high amounts of PFAS. As a result, several studies have been conducted to investigate the potential impact on drinking water in various areas along the river (Foguth et al., 2020; Kotlarz et al., 2020; Sun et al., 2016). The residents of Wilmington, North Carolina, depend primarily on the CFR as their main water supply. Additionally, the Cape Fear Public Utility Authority (CFPUA) provides water and sewage treatment to a population of >200,000 in New Hanover County, in southeastern North Carolina (Vandermeyden and Hagerty, 2020). From 1980 to 2017, a plant that produced fluorochemicals dumped wastewater into CFR containing PFAS. These PFAS included a number of fluoroethers, most notably HFPO-DA, often known as GenX (Kotlarz et al., 2020). Furthermore, CFR contained emerging PFAS such as perfluoro-3,5-dioxahexanoic acid (PFO2HxA), HFPO-DA (GenX), perfluoro-3,5,7-trioxaoctanoic acid (PFO3OA), perfluoro-3,5,7,9-tetraoxadecanoic acid (PFO4DA), perfluoro-2-methoxyacetic acid (PFMOAA) and Nafion byproducts (Guillette et al., 2020). Potential health concerns were mostly unclear (Vandermeyden and Hagerty, 2020). On June 15, 2022, the US EPA issued a final health advisory for GenX chemicals at 10 ng/L, given their potential toxicity to humans.

4.1. Methods used for detection of PFAS

Over the years, the US EPA has published EPA Method 537, 537.1, and 533 for measuring PFAS in drinking water (Table 2). In 2021, the EPA Draft Method 1633 for measuring PFAS in aqueous, solid, biosolids, and tissue samples was published. All these methods rely on using liquid chromatography-tandem mass spectrometry (LC-MS/MS) to measure targeted PFAS at ng/L levels. To identify PFAS with no known standards, suspect screening and non-targeted analysis can be performed using high-resolution LC-MS/MS. Additionally, the issues of real-time monitoring, cost-effectiveness, quick and accurate quantification, and a lack of skilled employees may be addressed via the use of analytical sensing, which is a feasible solution. Therefore, in recent years, different types of sensors, including nanomaterials-enabled, polymeric, optical fibrous, and metal-organic frameworks-based sensors, have been developed to measure PFAS fast and less costly (Garg et al., 2022; Menger et al., 2021; Rodriguez et al., 2020). Given the extremely low PFAS concentrations and their presence as highly complicated mixtures, the accuracy provided by these sensors still needs to be improved significantly.

4.2. Surface water contamination

PFAS have been discovered in aquatic environments, plants, and animals worldwide (Banzhaf et al., 2017). Early research of PFAS in the CFR basin indicated a widespread availability of PFAS across the river, likely originating from a variety of sources (Nakayama et al., 2007). Table 2 summarizes the distribution of PFAS across the state of North Carolina. For example, the Georgia branch was found to have the highest overall PFAS content. Furthermore, in another study, twelve new perfluoroalkyl ether carboxylic and sulfonic acids have also been found in the surface water of North Carolina. These compounds are made up of a series of similar perfluorinated and polyfluorinated substances with repeating CF_2 or CF_2O subunits (Strynar et al., 2015). In addition, a different study only discovered

Table 2
PFAS concentration in different locations in North Carolina (ng/L).

Location	Segment	Total ΣPFAS	PMPA	GenX	PFO2HxA	PEPA	PFMOAA	References
Georgia Branch	GW	2240	804	574	359	299	102	(Petre et al., 2021)
	SW	2484	895	629	386	286	136	
Willis Creek	GW	602	244	184	141	90	73	
	SW	593	246	96	95	60	27	
Mines Creek	GW	1546	639	355	196	195	77	
	SW	1511	542	407	215	177	71	
Kirks Mill Creek	GW	598	241	119	93	60	48	
	SW	–	–	–	–	–	–	
East of CFR 2	SW	726	376	85	78	80	44	
East of CFR 3	SW	1548	617	261	214	151	57	
East of CFR 4	SW	1062	442	158	156	102	43	

Location	Segment	ΣPFAS	PFPeA	PFNA	PFHxA	PFHpA	PFBS	PFOS	PFOA	PFBA	References
Haw River at the Bynum sampling station	SW	124.7	26.2	2	29.9	36.1	6.4	14.1	13.8	16.8	(Pétré et al., 2022)
CFR at Kings Bluff	SW	143	7.2	1.1	8.4	4.8	18.2	12.5	6.7	6.1	
Haw River at Bynum	SW	194	34	1.8	49.6	28	6.6	13.6	18.7	18.1	
Haw River Burlington Downstream	SW	219	40	1.8	38	18	8.7	9.8	9.7	21	
Haw River Burlington Upstream	SW	136	35	1.6	25	12	9.3	10	10	15	
Haw River Cane Creek 4	SW	192	–	–	–	–	–	–	–	–	
Jordan Lake	SW	68.5	–	–	–	–	–	–	–	–	
CFR Basin DWTP A	SW	355	44	<10	48	39	<10	44	46	33	(Sun et al., 2016)
CFR Basin DWTP B	SW	62	19	<10	<10	11	<10	<25	<10	12	
CFR Basin DWTP C	SW	710	30	<10	<10	<10	<10	<25	<10	22	

GW = Groundwater, SW = Stream water.

PFOA (8.07 ng/L) as well as PFHpA (5.79 ng/L) in Yadkin-Pee-Dee River water samples located in North Carolina (Penland et al., 2020). However, the EPA anticipates there are approximately 9000 different PFAS when the main PFAS and their metabolic and degradation products are added together (Baker and Knappe, 2022).

Recently, in Wilmington, North Carolina, several novel constitutional isomers were found in river water due to the increasing production of functionalized PFAS (Saleeby et al., 2021). In 83 % of the samples, constitutional isomers of $C_4H_2F_8O_4S_1$ were identified for the first time, and in 34 % of the water samples, a series of unique chlorinated PFAS was also detected (Saleeby et al., 2021). Furthermore, perfluoroalkyl ether carboxylic acids (PFECAs) are a type of fluorinated alternative that can be found in the CFR streams of a PFAS manufacturing plant. The PFECAs that have been identified include perfluoro-2-methoxyacetic acid (PFMOAA), perfluoro-4-methoxybutanoic acid (PFMOBA), perfluoro-3,5-dioxahexanoic) acid (PFO2H (PFO4DA), perfluoro-2-propoxypropanoic acid (PFPrOPrA), perfluoro-3-methoxypropanoic acid (PFMOPrA) (Sun et al., 2016). Consequently, three separate drinking water treatment plant (DWTP) water sources were found to contain legacy PFAS near the CFR Basin. The mean PFAS concentrations for these DWTP sites A, B, and C were 355, 62, and 710 ng/L, respectively (Sun et al., 2016). The total concentration differences between DWTP sites A and B could be described by the absence of major contributors to PFAS between A and B, dilution of streams, and the buffer activity of Jordan Lake, a major watershed between the two sites (Sun et al., 2016). The continued presence of PFAS in surface water in the CFR watershed means that as many as 1.5 million people may be at risk of being exposed to the chemicals (Pétré et al., 2022).

4.3. Groundwater contamination

Groundwater contamination by PFAS is significant because groundwater in many places of the world serves as a source of drinking water (Petre et al., 2021). As a result, humans and animals may be exposed to PFAS if they consume contaminated groundwater (Banzhaf et al., 2017). The groundwater of the CFR watershed in the central and coastal areas of North Carolina is heavily polluted by PFAS (Guillette et al., 2022). As a consequence, fluoroethers and legacy PFAS are still reaching the CFR in significant amounts, indicating groundwater release into the river and other outgoing inputs, despite the fact that the plant stopped discharging wastewater from the PFAS process in 2017 and PFOS and PFOA were banned from production and use in the USA in 2002 (Pétré et al., 2022). (Koropeckyj-Cox, 2019) examined PFAS-polluted groundwater beneath stream beds in the CFR basin near the PFAS manufacturing factory and quantified the flux of PFAS across groundwater to rivers through stream beds. Willis Creek had a groundwater-to-stream PFAS flux of 0.43 mg m^{-2} d^{-1}, while the Georgia tributary had a flux of 4.17 mg m^{-2} d^{-1}. It was also discovered that the five streams contributed 7.84 ng/L to the overall observed PFAS content in the CFR and 1.72 ng/L to the GenX concentration. Similarly, Petre et al. (2021) analyzed 29 PFAS, including perfluoroalkyl acids, from groundwater to CFR streams at Fayetteville Works in North Carolina, USA. Eight PFAS made up 98 % of the total PFAS; PMPA (perfluoro-2(perfluoromethoxy)propanoic acid), and GenX was 61 % of the total. The total PFAS concentration in groundwater ranged from 20 to 4773 ng/L, with an average of 1863 ng/L; the PFAS concentration in stream water, on the other hand, varied from 42 to 6361 ng/L, with an average of 1717 ng/L. There was an average of 32 kg/year of PFAS entering the CFR from groundwater in the five tributaries at baseflow. Thus, the release of contaminated groundwater could result in long-term pollution of surface water and adverse effects on drinking water supplies further downstream (Koropeckyj-Cox, 2019).

4.4. Air pollution

The Fayetteville Works facility in North Carolina has been emitting PFAS into the air since at least 1980. This release of PFAS has resulted in widespread contamination of the area's surface and groundwater (Gilmore et al., 2020). However, the impacts of air emissions on overall human intake are not sufficiently described. Recent research showed the air transit and distribution of a PFAS compound that originated from a North Carolina fluoropolymer manufacturing facility (D'Ambro et al., 2021). According to the community multiscale air quality model, 2.5 % of all GenX and 5 % of all PFAS emissions settled inside 150 km of the site. Overall, modeling showed that the air near the plant can have up to 24.6 ng/m^3 GenX and 8500 ng/m^3 of PFAS. These levels dropped to 0.1 and 10 ng/m^3 35 km away (D'Ambro et al., 2021). Average cumulative PFAS air concentrations around the facility reach a maximum of 8.5 μg/m^3 each year, and most of the emissions (about 95 % by mass) are carried >150 km away. In addition, PFAS masses upstream of the facility were found to be high, suggesting that the plant's direct water outflow is not the only source of PFAS in Cape Fear River (Sun et al., 2016). The amounts

found in the soil and nearby bodies of water, however, point to a possible role of air travel and deposition in the spread of pollution from the sites (Galloway et al., 2020). Furthermore, the potential risk of exposure by inhalation or other absorption routes is unknown, and the estimated atmospheric lifespan of PFAS could change significantly by the mass of PFAS emissions (D'Ambro et al., 2021). Separate research indicates that between 2012 and 2016, the fluorochemical production factory near Fayetteville, North Carolina, released between 226.8 and 303.9 kg of C_3 dimer acid fluoride into the environment, while the actual emissions may have been much higher (Hopkins et al., 2018). Similarly, airborne PFAS were recorded by (Zhou et al., 2022) on PM2.5 filters near the same major fluoropolymer production plant close to Fayetteville, North Carolina. There were 22 PFAS compounds: twelve emerging and ten legacies; five of each group were found to have the highest levels greater than one pg/m^3. Most of the overall PFAS content (86 %) was attributed to the following compounds: PFOS, PFBA, NVHOS, PFHxA, PFHxDA, Nafion BP1, PFO5DoA, and PMPA. Thus, airborne releases from production facilities and worldwide atmospheric transfer could be a source of contamination (Galloway et al., 2020), as PFAS are detectable even in distant regions where no direct PFAS discharges have occurred in a given watershed (Washington et al., 2019).

PFAS are found in many common household items, which can contribute to PFAS indoors and outdoors (Kotthoff et al., 2015). Some people may be exposed to PFAS through the dust and air in their homes (Poothong et al., 2020). For example, indoor dust was taken from 184 residences in North Carolina to investigate the availability of PFAS (Hall et al., 2020). The most common PFAS in household dust were fluorotelomer alcohols (FTOH) and dipolyfluoroalkyl phosphoric acid esters (diPAP), with average concentrations of at least 100 ng/g of dust. Furthermore, sorption to atmospheric aerosol particles allows the long-distance transfer of PFAS (Faust, 2023). The number of measurement sites for long-term observation of PFAS in particulate matter in North Carolina and beyond should be increased to understand the real impact.

4.5. Sediment contamination

Most pollutants originating from human activities end up in sediments. Therefore, evaluating the distribution of PFAS in sediment can provide information on how PFAS are stored and potentially released back into the water column (Balgooyen and Remucal, 2022). The long-term fate of PFAS in North Carolina sediments has not been well investigated. However, PFAS compounds have recently been identified in sediments taken near a direct influent line that supplies drinking water to a settlement located further downstream from the CFR. The sediment contained Nafion by-product 2 and two potential new isomers (Saleeby et al., 2021). The findings are significant because they show that benthic creatures and the food webs that depend on them are exposed to PFAS. Additionally, several PFAS compounds discovered in the CFR were also found in sediment collected from the same location, demonstrating interconnection between environmental compartments (Saleeby et al., 2021). Furthermore, over 12 weeks, the sedimentary fate of GenX was studied in southeastern North Carolina (Harfmann et al., 2021). Within 14 days, GenX in freshwater and estuary sediments decreased by 40–59 %, which could not be described by biological degradation because no degradation products were found. Therefore, water sediments could serve as a significant long-term sink for GenX as a result of their high sorption affinity and resistance to biological degradation.

The distribution of PFAS in sediment was recently determined by measuring their quantity along a CFR-to-ocean transect (Shimizu et al., 2022). The most prevalent PFAS chemicals were HFPO-DA, PFMOAA, PFOA, and PFOS, and their concentrations varied from under the LOQ (limit of quantification)- 7.47 ng/g dry weight. Perfluorinated ether sulfonic acid and perfluorinated ether carboxylic acid containing one or two acidic functional groups were also discovered in the upper CFR. Therefore, the subsequent release of PFAS from sediments into the surrounding water can also have a toxic effect on aquatic creatures and public health if the water body is a source of water supply (De Silva et al., 2021).

4.6. Drinking water contamination

Human exposure to PFAS can occur primarily through drinking water, regardless of whether the water comes from surface water or groundwater (Hopkins et al., 2018; Hu et al., 2016; Mulhern et al., 2022). It is estimated that >200 million people in the United States consume water that contains PFOS and PFOA in concentrations of 1 ng/L or higher. Additionally, the authors of the study estimate that between 18 and 80 million people in the United States consume water that contains PFOS and PFOA at concentrations of 10 ng/L or higher (Andrews and Naidenko, 2020). Although the EPA has announced interim health advisory level (HAL) for PFOA and PFOS of 0.004 ng/L and 0.02 ng/L, respectively, these HALs are not enforceable, which means suppliers of drinking water are not obligated to meet these levels (Hogue, 2022). Fig. 3 shows the path of contamination in the drinking water of North Carolina. North Carolina's drinking water sources have been found to contain various PFAS compounds, including several with known adverse health effects (Kluck et al., 2021). Fig. 2 (B) shows that all counties in North Carolina had total PFAS concentration levels over the HALs. The highest concentrations were observed in New Hanover, Brunswick, Pender, Bladen, Harnett, Moore, Guilford, Orange, Johnston, and Robeson counties. The map also shows that the highest PFAS concentrations in drinking water were found in the vicinity of the Chemours plant and the Cape Fear River watershed. For instance, next to a PFAS factory in North Carolina's spring-fed lakes, GenX concentrations as high as 4000 ng/L were found in private well water (Hopkins et al., 2018). Moreover, it is clear that counties like Harnett, Moore, Guilford, Orange, Johnston, Forsyth, and Davidson in the middle of North Carolina had higher PFAS concentrations. These fluoroethers were also not eliminated to a noticeable extent by the CFPUA's water treatment processes employing various advanced treatment methods (Hopkins et al., 2018; Zhang et al., 2019). Therefore, people living in Wilmington, North Carolina, may be vulnerable to fluoroethers in their drinking water (Rosen et al., 2022). Even though Fayetteville Works' direct PFAS wastewater discharge to the CFR was discontinued in 2017, the CFPUA has recorded a higher overall measurable PFAS content of approximately 377 ng/L in its untreated and processed water in 2019 (Petre et al., 2021).

The North Carolina PFAS Testing (PFAST) Network is a regional research program that was created to analyze present levels of PFAS compounds in water and airborne samples taken from several areas in North Carolina (PFAST, 2019). Throughout the state in 2019, they analyzed raw, untreated water from municipal surface water intakes and groundwater wells. Table 3 summarizes the total PFAS concentration of 18 different utility authorities in the major cities of North Carolina. For example, the total PFAS concentrations in the CFPUA and the Brunswick County water system were 406.9 and 395.4 ng/L, respectively. In addition, GenX and other fluoro-ethers were discovered in the drinkable water of the lower CFR, which was the primary route of exposure. The effects of exposure to these emerging PFAS on human health, however, remain little understood (Kotlarz et al., 2020). Therefore, in 2017, the *North Carolina* Department of Health and Human Services (NCDHHS) established a provisional health objective for GenX in drinking water at 140 ng/L (Roostaei et al., 2021; Vandermeyden and Hagerty, 2020).

5. Harmful impacts of PFAS on North Carolina residents' health

Growing concerns about the long-term effects of PFAS on the environment as well as public health, have emerged during the past 50 years, especially over the past decade (Baker and Knappe, 2022). Estimates show that many legacy PFAS have half-lives in humans of years, indicating that PFAS have alarming bioaccumulation and biomagnification potential (Baker and Knappe, 2022). Numerous harmful health risks, including impaired immunological function and chronic autoimmune diseases (Guillette et al., 2022), hepatotoxicity, cancer, decreased fertility, and toxic developmental effects, are associated with exposure to PFAS (Sunderland et al., 2019). For example, the consumption of locally captured freshwater fish contaminated with these PFAS chemicals has been linked to an increased risk of cancer

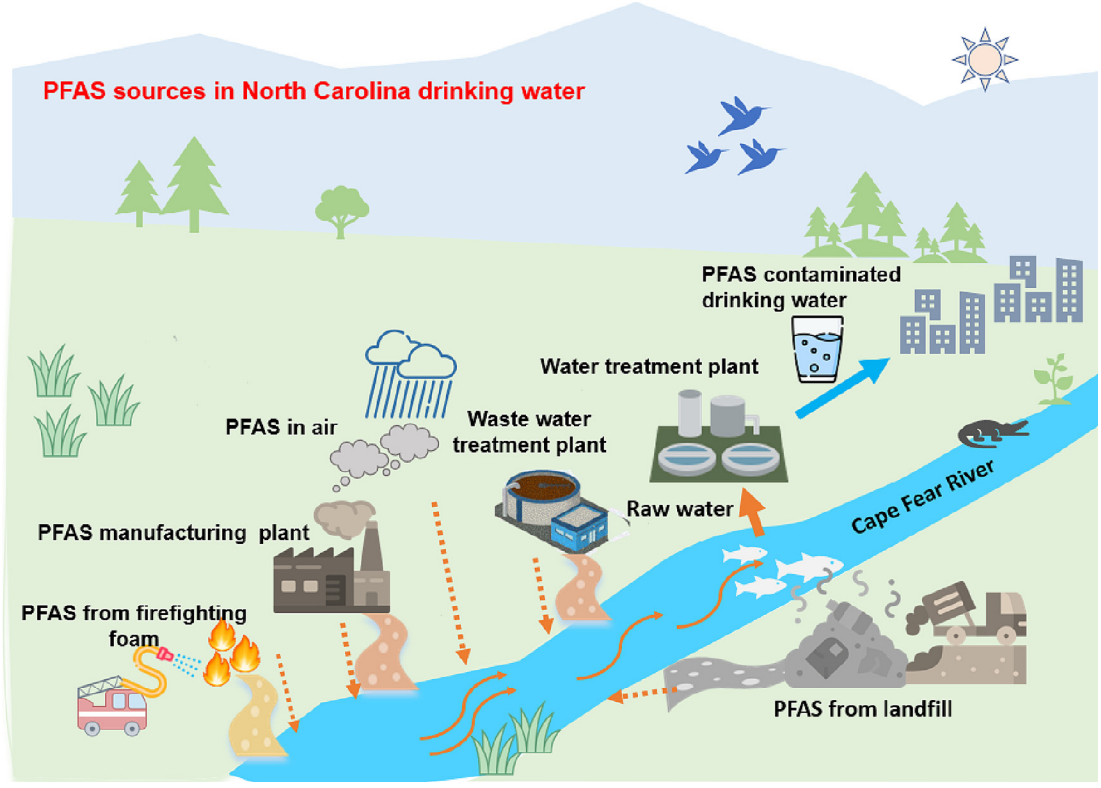

Fig. 3. A conceptual diagram describing drinking water contamination in North Carolina by emerging contaminant PFAS from different sources such as PFAS processing plants, firefighting foam, landfill leachate, and wastewater treatment plant.

and other health problems (Dwhall, 2023). In human blood serum, PFOA and PFOS are the most commonly found compounds (Olsen et al., 2007). Around 98 % of the population of the United States was found to have measurable amounts of one or more PFAS in their blood between 1999 and 2012 (EPA, 2016). The fluoroether, GenX, has been found in alarmingly high concentrations in the drinking water and the river water of Wilmington collected downstream of Fayetteville Works (Sun et al., 2016). Consequentially, (Kotlarz et al., 2020) tested serum samples from people in Wilmington to find out how much fluoroether and legacy PFAS were in them. >85 % of the respondents had detectable levels of PFO4DA, Nafion by-product 2, and PFO5DoA in their serum. PFO3OA and NVHOS, however, were rarely detected, while GenX was never detected at concentrations >2000 ng/L. Serum samples showed no measurable levels of GenX, despite the fact that the compound was still present in the water supply (at 50 ng/L). In addition, nearly all the respondents (97 %) had elevated concentrations of at least one of the four legacies PFAS tested: PFHxS, PFOA, PFOS, or PFNA. According to the 2015–2016 National Health and Nutrition Examination Survey, these concentrations were significantly higher. Recently (Rosen et al., 2022) looked at how serum fluoroether and legacy PFAS levels were related to different cholesterol outcomes. The participants were found to have geometric mean values of 8.08 ng/mL for PFOS and 4.13 ng/mL for PFOA. It was also observed that PFNA and PFOS were linked to increased levels of total and non-HDL cholesterol, with the relationships being stronger in the elderly. (Andersen et al., 2021) also observed human exposure to PFOA and PFOS is positively linked with elevated serum cholesterol levels.

Table 3
Total PFAS concentration of raw untreated water from municipal surface water intake in different locations in North Carolina (PFAST, 2019).

Utility authority	Total PFAS concentration (ng/L)	City	No. of people served
Brunswick county water system	395.4	Bolivia	100,694
CFPUA	406	Wilmington	153,202
Cary	95.6	Apex	192,250
Charlotte Water	6.8	Charlotte	1,093,901
City of Ashville	Not detected	Ashville	124,300
City of Concord	27	Concord	107,188
City of Durham	9.2	Durham	282,343
City of Greensboro	62.4	Greensboro	290,201
City of Highpoint	66.2	Highpoint	114,183
City of Raleigh	34	Raleigh	603,000
City of Winston Salem	66	Pfafftown	371,063
Davidson Water Inc.	18	Welcome	147,455
Fayetteville Public Works Commission	115.5	Fayetteville	217,948
Greenville Utilities Comm	23.2	Greenville	103,140
Harnett County Department of Public Utilities	47.6	Lillington	101,389
Onslow Water & Sewer Authority	1.6	Jacksonville	141,409
Piedmont Triad Regional	57.7	Randleman	367,681
Union County Water System	Not detected	Monroe	134,066

PFAS levels were also studied in the placenta and sociodemographic risk variables in a group of high-risk pregnant women in Chapel Hill, North Carolina (Bangma et al., 2020). Higher levels of PFUnA, PFHpS, PFHxS, and PFOS were found in 49, 55, 75, and 99 % of placentas, respectively, compared to the reporting limit. However, this study did not find a correlation between PFAS concentrations and hypertensive pregnancy disorders, fetal development, or gestational age. Likewise, in Durham, North Carolina, 120 placenta samples were collected after birth to determine the amount of PFAS in the placenta and examine any possible relation with neonatal outcomes (such as birth weight and length of pregnancy) (Hall et al., 2020). PFOS, PFOA, PFNA, and PFDA were the most prominent and commonly found PFAS (all >96 % detectable frequencies) in all placenta samples. It was revealed that high levels of PFOS exposure were related to decreased birth weight for gestational age in newborn males. Compared to the control group, birth weight was 13 % lower in the male group with the most exposure. On the other hand, it showed a rise in birth weight for newborn females, with the highest exposure group exhibiting an 11 % rise in birth weight of females (Hall et al., 2020).

PFAS are linked to adverse health consequences during pregnancy, birth, and later life (Baker and Knappe, 2022; Szilagyi et al., 2020). Fetal growth limitation, preeclampsia, diabetes in pregnancy, and obesity in children have all been linked to PFAS exposure. However, given the lack of a clear understanding of PFAS toxicity, better analytical assessments and monitoring are needed to determine how they affect human health (Baker and Knappe, 2022).

6. PFAS in fish and other biota

High concentrations of PFAS compounds have been found in both the surface and groundwater of the CFR watershed in central as well as coastal North Carolina (Guillette et al., 2022). Many PFAS dissolve well in water and stay in the environment for a long time. As a result, PFAS can travel large distances across marine and freshwater habitats (Banzhaf et al., 2017). Based on EPA statistics, Table 4 details where and how often PFAS have been found and which fish species have been impacted throughout the state of North Carolina (EWG, 2023a, 2023b). Moreover, blood samples collected from fish and other animals living in the CFR were found to contain increased amounts of PFAS (Guillette et al., 2022). It was reported that crocodiles from the CFR had higher blood PFAS levels than a sample group from the adjacent Lumber River area. Therefore, the correlation between higher exposure to PFAS and altered immune functions suggests that PFAS significantly change immunological activity in American alligators, resulting in an autoimmune-like disease. PFOS was found in all of the samples, with concentrations as high as 977 ng/mL in the serum of CFR Striped Bass (Guillette et al., 2020). Additionally, perfluorononanoic and perfluorodecanoic acids were found in all specimens, while perfluorohexanesulfonic acid was found in over 98 %. In addition, 48 % of the samples had HFPO-DA with a range of 240 to 5850 ng/L, and 78 % of the samples had Nafion byproduct 2. On the other hand, domestic stripe bass produced in well water under monitored aquatic environments had 40 times less PFAS than contaminated fish. Apart from these observed PFAS concentrations, the risks associated with consuming fish and seafood caught in PFAS-polluted waters remain unclear (Guillette et al., 2020).

In another study, the trophic transmission and accumulation of 14 PFAS in the Yadkin-Pee-Dee River food chain in North Carolina and South Carolina were evaluated (Penland et al., 2020). The presence of PFAS was confirmed by investigations of water, sediment, organic compounds, and aquatic species at all levels of the food web. In addition, biofilm, a microbial community consisting of microorganisms like bacteria, fungi, algae, and protozoa that form the basis of the aquatic food web, was also discovered to contain a high concentration of PFAS. The highest mean concentration for perfluorooctanoic acid was 463.73 ng/g in biofilm samples. The highest levels of PFAS found in isolated samples of aquatic insect and fish tissue were 1670.10 ng/g and 797.00 ng/g, respectively. Trophic magnification factors, or TMFs, demonstrated that different taxonomic groups accumulated PFAS chemicals in a variety of unique ways. Therefore, the TMFs for consumers in the Yadkin-Pee-Dee River ranged from 0.17 to 2.33 for PFBS, PFHpA, PFOA, PFOS, and PFDA. Additionally, (Robuck et al., 2020) investigated the presence of new PFAS in juvenile seabirds from the CFR Estuary. Three novel PFAS were found in chicks hatched in CFRE downstream of a fluoropolymer manufacturing facility, including perfluorinated ether sulfonic acid and two perfluorinated ether carboxylic acids, PFO4DA and PFO5DoDA. Increased amounts of PFAS have been observed in fish, birds, alligators, and individuals living near the Cape Fear River, indicating the potential for profound consequences on ecosystems as well as human beings from this contamination (Kotlarz et al., 2020; Robuck et al., 2020). As a result of insufficient research exploration, it is still unknown how PFAS pollution affects aquatic ecosystems, and we do not know the risks associated with consuming fish and seafood caught in contaminated waters (Guillette et al., 2020).

Table 4
Total PFAS* in freshwater fish in North Carolina (EWG, 2023a, 2023b).

Waterbody	Fish species	Sample taken year	Total PFAS concentration (ng/kg)
Catawba River	Largemouth bass	2013	19,090
Yadkin-Pee-Dee River	Spotted bass	2013	9154
Yadkin-Pee-Dee River	Channel catfish	2009	71,700
Cape Fear River	Channel catfish	2013	17,200
Cape Fear River	Blue catfish	2013	14,364
Neuse River	Largemouth bass	2009	30,200
Contentnea Creek	Largemouth bass	2013	6573
Roanoke River	Striped bass	2013	14,467
Chowan River	Largemouth bass	2013	13,220
Chowan River	Largemouth bass	2013	7411

Total PFAS* mainly includes PFUnA, PFDoDA, PFDA, and PFOS. Some places also include FOSA, PFHxA, PFPeA.

7. PFAS removal status in water treatment plants

The environmental working group lists >140 water utilities companies in North Carolina that serve over 10,000 people. The presence of PFAS is observed in almost all treatment facilities (EWG, 2021). The Cape Fear Public Utility Authority (CFPUA) was established in 2007 due to the consolidation of the water and wastewater services offered by the cities of Wilmington and New Hanover County into a single, financially independent organization (Vandermeyden and Hagerty, 2020). Most of the water used by the Sweeney Water Treatment Plant in North Carolina comes from the lower part of the CFR and parts of its service are also sourced from groundwater as well. In 2016, researchers discovered that the lower part of the CFR was contaminated with PFAS downstream of a fluorochemical-producing Chemours plant. This treatment plant serves approximately 200,000 customers (Jansen, 2019). This Plant (WTP) of CFPUA offers state-of-the-art treatment through pre-ozonation; coagulation; flocculation; clarification; intermediate ozonation; biologically active filtering; UV disinfection; stabilization; and chlorination (Vandermeyden and Hagerty, 2020). It is known that treatments with coagulation/sedimentation/filtration, chlorination/chloramination, ozonation and UV/H_2O_2 treatments are not effective for GenX, short- and long-chain PFAS (Hopkins et al., 2018). GAC adsorption, anion exchange, and high-pressure membranes (nanofiltration, reverse osmosis) are recognized as moderately to very effective for all three types of PFAS.

The latest tests of drinking water from the plant found only four PFAS, likely due to the installation of new Granular Activated Carbon (GAC) filters. The results of 69 different PFAS are documented in Table 5 (CFPUA, 2023a, 2023b). Among them, 65 were not detected in drinking water after treatment by the newly installed GAC filters. CFPUA used a modified EPA method 537.1, with isotope dilution. The experimental detection limit for PFAS varies by compound from 0.587 ng/L to 1.58 ng/L. Therefore, analytical results that did not meet these limits were reported as “ND” by the laboratory. Therefore, it was not able to verify whether the PFAS concentrations reported as “ND” complied with health advisory standards published in June 2022 (PFOA and PFOS, 0.004 and 0.02 ng/L, respectively). The brand-new filters at the Sweeney WTP require nearly 3 million lbs. of GAC, making it the largest GAC water treatment facility in North Carolina. A total of $43 million was spent on the development and construction to improve the treatment process. Operational expenses will total $3.7 million in the fiscal year 2023, rising to $5 million in the following years (CFPUA, 2022). Similarly, the Northwest Water Treatment Plant in Brunswick County just completed a $137 million reverse osmosis project in 2022 to remove PFAS (Stoiber et al., 2020). Brunswick and New Hanover counties are the most affected in North Carolina, according to Fig. 2(B). Charlotte Water is another large utility company that serves >1 million people in North Carolina. On March 1st, 2023, the most recent results of Charlotte water showed that the PFOS and PFOA content in drinking water was 3.6 and 1.9 ng/L, respectively (Water, 2023). The Town of Cary tested 39 PFAS in the drinking water multiple times in 2020. Seven PFAS were found. Compared to the US EPA recommended health advisory level of 70 ng/L, the total PFOA and PFOS ranged from 3.2 to 6.7 ng/L at Cary (Revels, 2020).

Furthermore, North Carolina has the second-highest overall population (3.3 million) of residents who rely on private wells for drinking the water

Table 5
Finished water testing results from the Sweeney Water Treatment Plant, Wilmington, North Carolina, reported on 1/10/2023 (CFPUA, 2023a, 2023b).

PFAS compound	Result (ng/L)	PFAS Compound	Result (ng/L)
Perfluoro(2-ethoxyethane) sulfonic acid (PFEESA)	ND	Perfluoroheptanesulfonate (PFHpS)	ND
Fluorotelomer sulfonate 10:2 (10:2 FTS)	ND	Perfluorononanoic acid (PFNA)	ND
PMPA	1.47	Perfluorotetradecanoic acid (PFTeDA)	ND
2,3,3,3-Tetrafluoro-2-(1,1,2,2,3,3,3-heptafluoropropoxy)-propanoic acid (PFPrOPrA) GenX	ND	Sodium 2,2,4,4,6,6,8,8,10,10,12,12,12-tridecafluoro-3,5,7,9,11-pentaoxadodecanoate (PFO5DA)	ND
Nonafluoro-3,6-dioxaheptanoic acid (NFDHA)	ND	Perfluoro-3-methoxypropanoic acid (PFMOPrA or PFMPA)	ND
Perfluorooctadecanoic acid (PFODA)	ND	Perfluoro(3,5-dioxahexanoic) acid (PFO2HxA)	ND
2-(N-ethylperfluoro-1-octanesulfonamido)-ethanol (N-EtFOSE)	ND	Perfluoro(3,5,7,9-tetraoxadecanoic) acid (PFO4DA)	ND
Perfluorooctanesulfonate (PFOS)	ND	Perfluoro(3,5,7-trioxaoctanoic) acid (PFO3OA)	ND
Perfluoroundecanoic acid (PFUdA)	ND	Fluorotelomer sulfonate 8:2 (8:2 FTS)	ND
N-methylperfluoro-1-octanesulfonamidoacetic acid	ND	N-ethylperfluoro-1-octanesulfonamide (N-EtFOSA)	ND
Byproduct 4 (BP4); R-PSDA	ND	Perfluoropropanoic acid (PPF Acid) PFPrA	5.86
Byproduct 5 (BP5); Hydrolyzed PSDA	ND	Perfluorohexadecanoic acid (PFHxDA)	ND
Byproduct 6 (BP6); R-PSDCA	ND	Perfluorononanesulfonate (PFNS)	ND
R-EVE	ND	Perfluoro-2-methoxyacetic acid (PFMOAA)	1.67
2-(*N*-methylperfluoro-1-octanesulfonamido)-ethanol (N-MeFOSE)	ND	2-Perfluorohexyl ethanoic acid (6:2 FTA) (FHEA)	ND
Sodium 2,3,3,3-tetrafluoro-2-(perfluoroethoxy)propanoic acid (PEPA)	ND	2-Perfluorodecyl ethanoic acid (10:2 FTA) (FDEA)	ND
Perfluoropentanoic acid (PFPeA)	ND	Perfluoroethoxypropionic acid (EVE Acid)	ND
Perfluoropentanesulfonate (PFPeS)	ND	Perfluorotridecanoic acid (PFTrDA)	ND
Fluorotelomer sulfonate 6:2 (6:2 FTS)	ND	Nafion Byproduct 2	ND
2-Perfluorooctyl ethanoic acid (8:2 FTA) (FOEA)	ND	9-chlorohexadecafluoro-3-oxanonane-1-sulfonate	ND
Nafion Byproduct 1	ND	Perfluorooctanesulfonamide (PFOSA)	ND
N-ethylperfluoro-1-octanesulfonamidoacetic acid	ND	11-chloroeicosafluoro-3-oxaundecane-1-sulfonate (PF3OUdS)	ND
FBSA	ND	Fluorotelomer sulfonate 4:2 (4:2 FTS)	ND
Perfluorohexanoic acid (PFHxA)	ND	Hydro-EVE	ND
Perfluorododecanoic acid (PFDoA)	ND	Perfluoro-1-dodecanesulfonate (PFDoS)	ND
N-methylperfluoro-1-octanesulfonamide (N-MeFOSA)	ND	1,1,2,2-Tetrafluoro-2-(1,2,2,2-tetrafluoroethoxy)ethane sulfonic acid (NVHOS)	ND
Perfluorooctanoic acid (PFOA)	ND	Perfluorodecanesulfonate (PFDS)	ND
Perfluorodecanoic acid (PFDA)	ND	7:3 FTCA	ND
4-(Heptafluoroisopropoxy)hexafluorobutanoic acid (PFECA-G)	ND	Perfluoro-4-methoxybutanic acid (PFMOBA or PFMBA)	ND
Perfluorohexanesulfonate (PFHxS)	ND	5:3 FTCA	ND
4,4,5,5,6,6-Heptafluorohexanoic acid (3:3 FTCA)	ND	Sodium dodecafluoro-3H-4,8-dioxanonanoate (ADONA)	ND
Perfluorobutyric acid (PFBA)	0.95	NVHOS	ND
Perfluorobutanesulfonate (PFBS)	ND	N-CMAmP-6:2 FOSA	ND
Perfluoroheptanoic acid (PFHpA)	ND	FHxSA	ND
N-AP-FHxSA	ND		

ND = not detected.

behind Pennsylvania (Maupin et al., 2014). Eighteen consumers of private wells were contacted to evaluate the efficacy of under-sink activated carbon block water filters in removing PFAS (Mulhern et al., 2022). The filters removed 97 % to 99 % of 17 PFAS (Σ PFAS 4.7–131 ng/L), along with short-chain PFEA, that were in the water. Therefore, PFAS can be removed from domestic well water using filters that have National Sanitation Foundation international certification NSF P-473 to meet the standards set forth in the Public Health Guidelines. In addition, the efficacy of domestic point of entry (POE) and point of use (POU) water filters were evaluated for eliminating 16 distinct PFAS from central and southeastern North Carolina homes (Herkert et al., 2020). Most of the PFAS were eliminated after being filtered using reverse osmosis systems installed beneath the sink. All other activated carbon filters removed PFAS to different extents. These filters were more effective (60–70 %) at removing long-chain PFAS than short-chain PFAS (40–60 %) from drinking water.

8. Socioeconomic impacts of the affected community of North Carolina

People are becoming more aware of the harm that could come from being exposed to PFAS compounds (B.Cahoon, 2020). Early in June 2017, Wilmington residents learned that GenX was present in local drinking water (Kotlarz et al., 2020). For those who rely on private well water, technical, educational, and economic assistance is strongly needed to ensure the drinking water is PFAS-free (Mulhern et al., 2022). After GenX and related perfluoroalkyl ether acids (PFEA) were discovered in significant amounts in finished drinking water, a swift response from legislators, health authorities, commercial testing agencies, water distributors, and professional consultants was kicked off (Hopkins et al., 2018). Subsequently, Chemours and DuPont were sued by CFPUA in October 2017 in the federal district court for the Eastern District of North Carolina. For decades, Chemours and DuPont released PFAS compounds in CFR, such as GenX and others, from their chemical factory on the Bladen-Cumberland county boundary, nearly 100 miles upstream from Wilmington, triggering ongoing litigation (CFPUA, 2023a, 2023b). Additionally, a regional environmental organization filed a civil action under the Clean Water Act against Chemours in February 2019 (Vandermeyden and Hagerty, 2020).

People living in PFAS-contaminated areas struggle with the anxiety of not knowing how contamination may affect their health and their families. Inaccuracies and discrepancies clouded their knowledge of the contaminants they encountered. However, CFPUA has organized several community forums to address consumer concerns and share the findings of its PFAS monitoring (Vandermeyden and Hagerty, 2020). As a result of a lack of health data and inconsistent interpretations of risk about the health implications of exposure to PFAS, affected residents were left to navigate various sources of information to know about their vulnerabilities (Wickham and Shriver, 2021). Immediate steps must be taken to examine and improve official health messages that are designed to educate the general public and medical professionals about the dangers of exposure to PFAS (Ducatman et al., 2022).

9. Implications for better control of PFAS

Widespread usage of now-banned or -replaced PFAS, especially PFOS, is nevertheless frequently detected at AFFF-impacted sites, posing ongoing dangers to the environment (Reinikainen et al., 2022). When it comes to health and the environment, PFAS migration into aquatic ecosystems, fish intake, and groundwater use poses the greatest concerns near military

sites or commercial airports. As a result, the North Carolina General Assembly has mandated that by the end of 2019, the North Carolina Policy Collaboratory at the University of North Carolina at Chapel Hill will have compiled a comprehensive database of all AFFF used or stored by North Carolina's municipal fire departments, including those at or near airports (Bodnar, 2021). The team suggested that performance criteria and system requirements for firefighting foam should be considered, and processes should be constructed with containment control, runoff control, and foam (and unburned fuels) collection to prevent contamination of neighboring rivers and soil. AFFF containing short chain PFAS (C6 or fewer) or fluorine-free foams may be used in place of long-chain AFFF in situations where there is an imminent danger to human life, public safety, or property. Furthermore, manufacturers now offer fluorine-free alternatives to AFFF containing PFAS, which should be used in all firefighting exercises. In North Carolina, fire departments must address a lack of knowledge and information on the use of AFFF and the significant risk of PFAS exposure.

Given the fact that PFAS are discharged from different sources and multiple channels, source apportionment should be assessed to understand the relative contribution of the various types of PFAS to the environment. Furthermore, it is necessary to identify sources and assess the contribution and severity of individual sources to create and prioritize implementing effective mitigation solutions for the elimination or reduction of PFAS in North Carolina. A promise by the industry to share data on the chemical compositions and analytical procedures of newly commercialized PFAS would be an example of increased transparency (De Silva et al., 2021). Furthermore, extensive research is needed to understand the distribution of PFAS in the soil/sediment around the area affected by PFAS in order to plan a remediation strategy to minimize the long-term effects.

For the point of entry (POE) and point of use (POU), installing a reverse osmosis system beneath the sink or using a granulated activated carbon (GAC) system are the two best ways for homes to lower the amount of PFAS in their drinking water. Moreover, strategies to reduce exposure to PFAS include following local consumer guidelines for game and fish, using cast-iron kitchenware instead of Nonstick pans, avoiding stain-resistant fabric, and avoiding weatherproof sprays and products containing PTFE (Alan D. Woolf and Zajac, 2022).

10. Future challenges

New per and polyfluorinated species have been developed in response to the restriction of legacy PFAS, such as PFOA and PFOS (McCord and Strynar, 2019). Additionally, long-chain PFAS production has been cut back or discontinued in the United States because of their ability to remain in the environment, accumulate in organisms, and cause harm (Woodlief et al., 2021). Through the EPA's voluntary stewardship program, PFOA substances were eventually phased out and replaced by GenX (Chemours, 2017). The lack of essential standards and the limited knowledge of replacement chemistries make screening and monitoring these new chemicals difficult. However, the environmental stability of these perfluorinated compounds with shorter chains is expected to be the same as that of the associated longer-chain acids (Baker and Knappe, 2022; Strynar et al., 2015). One of the most concerning issues is that some legacy PFAS have been discovered in samples dating back to the 1950s, indicating that they are currently present after 70 years (Baker and Knappe, 2022).

The majority of traditional and cutting-edge treatment methods face challenges in removing legacy PFAS (Appleman et al., 2014; Sun et al., 2016). With respect to removing PFAS in the CFR, in addition to deep-bed granular activated carbon (GAC) (CFPUA, 2022), additional technologies such as ion exchange (IX) or low-pressure reverse osmosis (RO) may be required to reduce PFAS to satisfactory levels (Vandermeyden and Hagerty, 2020). Aside from technology barriers to completely eliminating PFAS from the water, the lack of knowledge and skills regarding the installation and maintenance of POU treatment posed substantial difficulties in reaching water consumers (Mulhern et al., 2022). This is worsened by the fact that up to 1.5 million individuals in North Carolina could potentially be exposed to PFAS and would require suitable technologies to remove PFAS from their drinking water (Pétré et al., 2022).

The health consequences of exposure to legacy and emerging PFAS must be thoroughly defined. Moreover, human exposure to fluorochemical manufacturing by-products also must be examined (Kotlarz et al., 2020). The responsible recycling of fluoropolymers and related industries and materials at the end of their useful life is also an issue of concern (Lohmann et al., 2020). There are not many ways to recycle them from consumer products. Toxic air pollutants (PFAS) in dust pose a severe problem, so tracking their origins is crucial. More research is needed to identify the many dust sources that may include PFAS (Hall et al., 2020).

11. Conclusions

North Carolina (NC) is one of the most affected states in the United States, with >10 million people living. Almost every county in North Carolina has PFAS contamination. This environmental and geographical pervasiveness of PFAS poses risks to environmental health, as numerous legacy PFAS and newly developed PFAS are harmful to many organisms. The results of this review of North Carolina revealed the following.

1. Drinking water sources in North Carolina, like CFPUA and the Brunswick County water system, have been discovered to contain total PFAS of 406.9 and 395.4 ng/L, respectively. Therefore, several North Carolina rivers, including the Cape Fear River, have been found to contain high amounts of PFAS that significantly impact the reliability of drinking water of North Carolina residents.
2. Sources of PFAS in North Carolina are concentrated on four major segments: large fluoropolymer production facilities in Fayetteville, firefighting foam, landfill leachate, and wastewater treatment plant. Maximum concentrations detected in identified sources of PFAS range from 70 to 312,000 ng/L between 2017 and 2020, and PFOS, PFOA, and GenX are significant PFAS compounds.
3. It is known that there are a significant number of PFAS that accumulate in groundwater, surface water, wildlife, and aquatic species at all levels of the food chain, including bacteria, fungi, algae, protozoa, fish, birds, and alligators. The total concentration of PFAS found in surface and groundwater in North Carolina ranges between 62 and 2484 ng/L. Furthermore, the PFAS found in freshwater fish are between 6573 and 71,700 ng/kg.
4. It was found that PFAS levels varied considerably between food web segments and fish species. Plant samples were the least likely to have PFAS found in them (16 %). High PFAS deposition was seen in biofilm, an aquatic food web foundational resource consisting of bacteria, fungus, algae, and protozoa. The greatest levels of PFAS were noticed in aquatic insects compared to other taxa. This suggests a trophic relationship involving biofilm PFAS with aquatic insect PFAS. Fish showed varying PFAS detections depending on the species.
5. Granular activated carbon (GAC), high-pressure membrane filtration, and ion-exchange resin are all effective strategies for removing some PFAS. Recently, the Sweeney Water Treatment Plant in Wilmington introduced GAC treatment and removed a substantial amount of targeted PFAS (Table 5). However, due to limitations in the PFAS testing instrument, analytical challenges exist to detect PFAS concentrations lower than the detection limit. Water treatment facilities in North Carolina should perform a case-by-case assessment of available treatment techniques before deciding which ones to implement to eliminate PFAS.
6. >85 % of Wilmington, North Carolina, respondents had PFAS in their serum (Kotlarz et al., 2020), and there was evidence that it had an adverse effect on birth weight. However, more extensive research is required to evaluate the impact of PFAS exposure on North Carolina's public health.
7. More research is needed to determine the effects of exposure to emerging PFAS on human health, the environmental fate of PFAS, the development of a standard for monitoring and screening of new PFAS both at the regional and national scale, and the methods for the safe recycling and disposal of fluoropolymers.

The effects of PFAS on the ecosystem of North Carolina are the subject of this intensive study. However, there is still a lot of work to do to gain a deep understanding of the actual state of North Carolina. The state needs to continue monitoring, research, and action to mitigate the risks associated with PFAS contamination, including identifying and tracking sources of contamination, implementing effective treatment strategies, and regulating and restricting the use of PFAS in commercial and industrial products. In addition, the community must realize that future generations will use the same polluted environment we live in now. Therefore, ensuring a clean and healthy ecosystem and a reliable supply of safe drinking water is vital.

CRediT authorship contribution statement

Conceptualization, M.N.E. and M.R.; formal analysis, M.R. M.N.E. M.N.P. Y.L. and M.M.O.K.; writing—original draft and final paper preparation, Y.L. and V.N.; writing—review and editing, M.R. M.N.E. and V.N.; project administration, funding acquisition and supervision; All authors have read and agreed to the published version of the manuscript.

Data availability

Data will be made available on request.

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.

Acknowledgments

We would like to express our sincere gratitude for the support from the Sanitary Environmental Engineering Division (SEED) and grants (FARB projects) from the University of Salerno, Italy, coordinated by V. Naddeo. Grant Number: 300393FRB22NADDE. In addition, Y. Liang acknowledges funding support from the U.S. National Science Foundation under award number CBET 2225596.

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