

PFAS in drinking water: an emergent water quality threat

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PFAS (per- and polyfluorinated substances), drinking water, wastewater

Per- and polyfluoroalkyl substances (PFAS) gained notoriety as environmentally persistent contaminants detected in drinking water, air, food and the blood of nearly all people. Testing commissioned by Environmental Working Group, a U.S.-based nonprofit organization, found PFAS in the drinking water of dozens of U.S. cities, including major metropolitan areas. Research from around the world continuously discovers new areas with PFAS water contamination, primarily stemming from industrial wastewater discharges and the use of PFAS-based fire-fighting foam. As the full scope of the PFAS contamination is revealed, numerous communities are implementing ways to remove PFAS from their drinking water, through methods such as treatment with granular activated carbon, ion exchange or reverse osmosis. Example costs for PFAS removal from drinking water are reviewed here.

Introduction

Per- and polyfluoroalkyl substances (PFAS) are a group of synthetic, fluorinated chemicals that have been widely used in industrial and consumer products due to their water- and grease-repelling properties. PFAS are highly persistent contaminants that can accumulate in the environment and living organisms. There are thousands of PFAS chemicals, with more than 600 compounds in commercial use in the United States. PFAS chemicals are typically present in complex mixtures within consumer products and the environment. PFAS have been detected in people [1], including perfluorooctanoic acid (PFOA) and perfluorooctanesulfonate (PFOS), compounds with an eight-carbon fluorinated chain that are no longer produced in many countries and are restricted by the Stockholm Convention on Persistent Organic Pollutants [2]. Perfluorohexanoic acid (six fluorinated carbons), perfluorobutane sulfonic acid (four fluorinated carbons), and hexafluoropropylene oxide dimer acid, also known as GenX, a substitute fluorochemical introduced in place of PFOA, are also detected in people [1].

In epidemiological studies, exposure to PFAS, particularly PFOA and PFOS, is associated with changes in the hormonal balance and thyroid function, weakened immune response, increased cholesterol, harm to the developing fetus, and elevated risk of cancer [3, 4]. Yet, for most PFAS chemicals, no toxicity data are publicly available. In the absence of comprehensive information for the majority of PFAS, government agencies around the world are struggling to establish health protective policies to address PFAS contamination. For example, the U.S. En-

vironmental Protection Agency has published a lifetime health advisory of 70 ng/l (also described as parts per trillion, or ppt) for PFOA and PFOS individually or combined, while much lower guidelines for PFAS in drinking water have been published by state agencies in New Jersey and other U.S. states, and more stringent exposure recommendations for PFOA and PFOS have been developed by the U.S. Agency for Toxic Substances and Disease Registry [3]. Recent scientific analysis suggests that the exposure limit for PFOA and PFOS should be at or below 1 ng/l [5, 6].

Drinking water is the most significant source of PFAS exposure for communities whose water is impacted by industrial discharges from fluorochemical production, PFAS-based firefighting foam, and municipal wastewater treatment plant effluent [7-10]. Food represents another important route of exposure [11]. PFAS absorb into food from contaminated soil and water, PFAS pollution in the environment, and from food packaging where PFAS substances are used to impart grease- and water-resistance. Everyday products in the home, such as dental floss and PFAS-based carpet and furniture coatings, also contribute to PFAS exposure [11].

Detection of PFAS in drinking water

In 2013-2015, large community water systems in the U.S. tested their water for six PFAS chemicals, including PFOA and PFOS, in a program mandated by the U.S. Environmental Protection Agency. Due to the relatively high analytical reporting limits, limited number of PFAS chemicals tested, and the lack of water sampling in smaller commu-

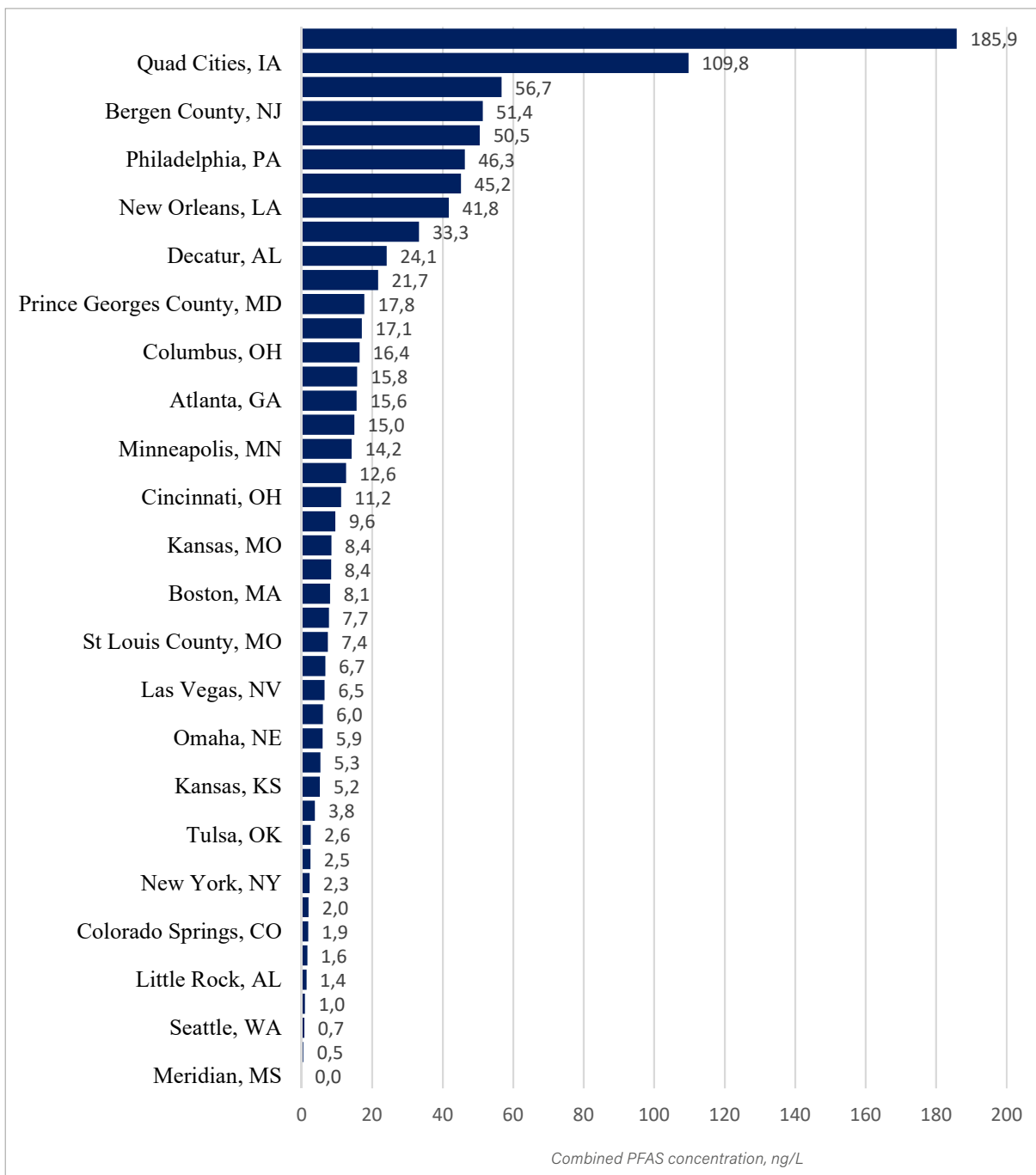


Figure 1: Combined PFAS concentrations (figure from Evans et al. [14]).

nities, this testing program underreported the extent of PFAS contamination [12]. Nevertheless, the testing results documented a correlation between the number of nearby industrial sites that manufacture or use PFAS compounds, the number of military fire training areas and the number of wastewater treatment plants with the frequency of detection of PFAS in public water supplies [13]. In 2019, laboratory tests commissioned by Environmental Working Group, a U.S.-based nonprofit organization, have for the first time reported the presence of PFAS in the drinking water of dozens of U.S. cities serving popu-

lations of different sizes and located in different regions, as shown in **Figure 1** and **Table 1** [14]. One sample was collected at each location and all samples were analyzed using a modified version of U.S. EPA method 537. For the 30 PFAS compounds included in the analysis, detection limits ranged from 0.3 to 2.0 ng/l, quantification limits ranged from 2 to 5 ng/l, and the total PFAS concentration was calculated as the sum of all detected compounds. The combined PFAS concentrations ranged from less than 1 ng/l to 185 ng/l [14]. Overall, these results indicate that the number of Americans exposed to PFAS

Table 1: PFAS chemicals detected in study by Evans et al. [14].

Compound	Abbreviation	Detection Limit	Percent of samples with detections	Range of detections, ng/l*
6:2 Fluorotelomersulfonic acid	6:2 FTSA	1	5	2.1-15
Perfluorooctanesulfonamide	FOSA	0.5	48	0.44-1.9
Hexafluoropropylene oxide dimer acid	GenX	0.5	14	0.5-31
Perfluorobutanoic acid	PFBA	2	73	1.8-72
Perfluorobutanesulfonic acid	PFBS	0.3	61	0.48-5
Perfluorodecanoic acid	PFDA	0.9	7	0.5-0.89
Perfluoroheptanoic acid	PFHpA	0.4	59	0.47-24
Perfluorohexanoic acid	PFHxA	0.4	70	0.44-36
Perfluorohexanesulfonic acid	PFHxS	0.4	52	0.45-7.3
Perfluorononanoic acid	PFNA	0.4	23	0.45-1.9
Perfluorooctanoic acid	PFOA	0.3	68	0.35-14
Perfluorooctanesulfonic acid	PFOS	0.4	77	0.43-14
Perfluoropentanoic acid	PFPeA	2	70	0.47-39
Perfluoropentanesulfonate	PFPeS	0.4	7	0.4-1.6

* Range of detections in samples where compound was found above the detection limit.

from contaminated tap water is much greater than what the U.S. EPA testing program showed.

Of tap water samples from 44 locations in 31 states and the District of Columbia, one site had no detectable PFAS and two other sites had a total PFAS concentration below 1 ng/l. In twenty locations all PFAS detects were above the laboratory limit of detection, although below the limit of quantification. Some of the highest PFAS levels detected were in samples from major metropolitan areas, including Miami, Philadelphia, New Orleans and the suburbs of New York City. In 34 places where this study found PFAS, contamination has not been publicly reported by the U.S. Environmental Protection Agency or state government agencies.

Two-dimensional hierarchical clustering identified distinct groupings of locations and PFAS profiles (**Figure 2**). Locations with greater levels of contaminants featured mixtures with a grouping of five to seven carbon PFAS as well as a grouping of the long-chain PFAS (PFNA, PFOA, PFOS) that are phased out in the U.S. Four-carbon chain compound PFBA was broadly detected in many areas. GenX was detected near the industrial release source in North Carolina, as well as in Cincinnati, Ohio; Louisville, Kentucky; and New Orleans, Louisiana. The variations in the PFAS profiles likely indicate distinct sources of contamination. Additional sampling and regional analysis will be required to identify the specific sources of contamination and their respective PFAS signatures.

As noted above, drinking water contamination with PFAS is typically due to local PFAS contamination sites or to ongoing PFAS discharges into the waterways. Environmental Working Group has been publishing documents and reports detailing PFAS contamination in the U.S. since 2002. A map of the available information for the U.S.-wide PFAS contamination is shown in **Figure 3** and available at www.ewg.org/interactive-maps/2019_pfas_contamination/map/. As of February 2020, the map includes nearly 1,400 locations with known contamination. The map compiles drinking water and ground water test results from federal and state testing programs and the currently released information about PFAS in drinking water and groundwater on and around military facilities and installations maintained by the U.S. Department of Defense. A research project conducted by U.S. Geological Survey and U.S. Environmental Protection Agency scientists analyzed PFAS levels in source water supplies and treated drinking water in 25 different water systems in the U.S. [15]. PFAS chemicals were detected in the water before and after treatment at every location and individual PFAS concentrations ranged from less than 1 ng/l (trace levels) to 1,102 ng/l. The number of PFAS detected in the finished drinking water ranged from 4 to 12 compounds, with a median concentration of 19.5 ng/l. The source water PFAS concentration matched the treated drinking water concentration for 23 out of 24 utilities that had more than trace levels of PFAS, indicating that con-

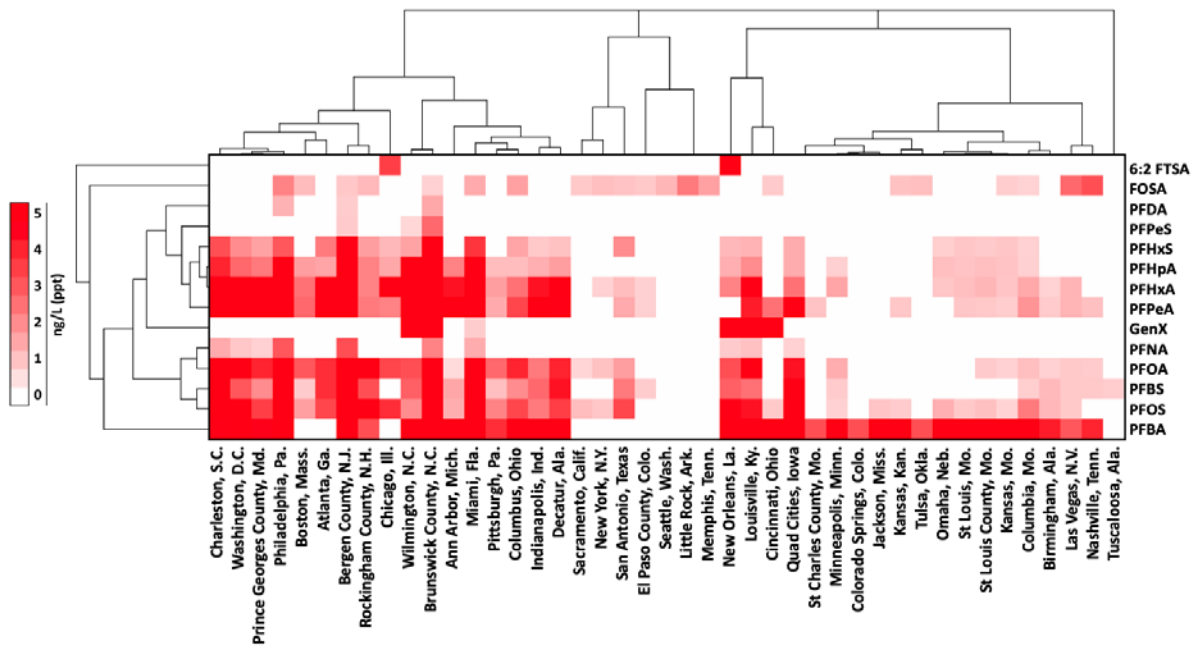


Figure 2: Relationships between detected PFAS and test site locations [14]. Two-dimensional hierarchical clustering (complete linkage using correlation (uncentered) as the similarity metric) was carried out using Gene Cluster 3.0 software. Results were visualized using Treeview v1.0.9 (<http://jtreeview.sourceforge.net>). The PFAS that were not detected in any city as well as the city of Meridian, Mississippi (where no PFAS were detected), were not included in the analysis. Tuscaloosa, Alabama was the only site with just one PFAS detection (PFBS).

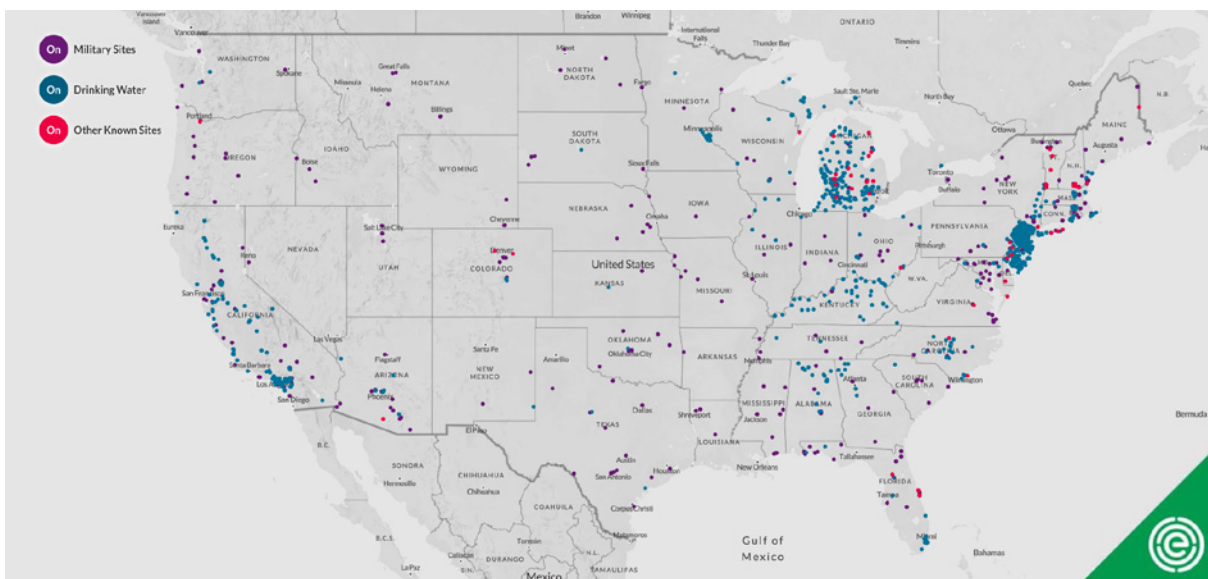


Figure 3: Map of PFAS detections in drinking water and groundwater across the United States, available at www.ewg.org/interactive-maps/2019_pfas_contamination/map/. Color code: purple – military sites with reported PFAS contamination in groundwater; blue – communities with PFAS water contamination; red – other known PFAS contamination sites.

ventional drinking water treatment technologies at those treatment plants could not reduce PFAS levels. Only one water system had a treatment train able to reduce both the number of PFAS compounds between source and treated water (from 12 to 6) and their combined concentration by approximately seven-fold [15]. Among the

water systems in the study, this specific system had the largest Granular Activated Carbon (GAC) bed depth and replaced the GAC most frequently, factors which likely contributed to its ability to remove PFAS. In Europe, high levels of PFAS in drinking water were reported in the Veneto region in Italy, caused by in-

Table 2: Example costs of drinking water treatment for PFAS in U.S. community water systems.

Location and reference for the cost estimates	Type	Installation Cost (as listed for the project)	Operating Cost and Capacity
Wilmington, North Carolina ¹	Plant upgrades and eight new granular activated carbon filters, 10,250 cubic feet of media at Sweeney Water Treatment Plant, operational 2022	\$46 million	\$2.9 million for capacity of 44 million gallons per day
Plainfield Township, Michigan ²	Five new GAC filters	\$750,000	Capacity, 12 million gallons per day
Ann Arbor, Michigan ³	New GAC filters, 11,000 cubic feet of media over two years	\$1 million in 2018, \$950,000 in 2019	Capacity, average 14 million gallons per day
Brunswick County, North Carolina ⁴	Reverse Osmosis upgrades to Northwest Water Treatment Plant, completed 2022	\$137 million	\$4.7 million annual at a flow of 16 million gallons per day
Decatur, Alabama ⁵	New Reverse Osmosis plant for West Morgan- East Lawrence Water Authority, completed 2020	\$30 million	\$1 million to operate at 10 million gallons per day capacity
Ridgewood, New Jersey ⁶	New GAC filters at Carr Treatment Plant	\$3.5 million	1 million gallons per day

¹ Cost estimates from Cape Fear Public Utility Authority <https://www.cfpu.org>

² Cost estimates from Plainfield Township <https://plainfieldmi.org>

³ Cost estimates from the city of Ann Arbor <https://A2gov.org> and from <https://mlive.com>

⁴ Cost estimates from engineering estimates developed by CDM Smith for advanced treatment options for the Northwest Water Treatment Plant, available at <https://brunswickcountync.gov> 2018

⁵ Cost estimates from West Morgan-East Lawrence Water and Sewer Authority <https://wml.org> and from <https://tennesseeriverkeeper.org> and <https://pfasproject.org>

⁶ Cost estimates from Ridgewood Water <https://water.ridgewoodnj.net> and from <https://northjersey.com>

dustrial waste discharges [16]. PFAS water pollution has been reported in Austria, Denmark, France, Germany, the Netherlands and Sweden [17]. Given the widespread nature of PFAS contamination in the environment and the variety of sources and pathways by which PFAS chemicals enter surface- and groundwater, it is probable that PFAS are present in drinking water in all European countries, and across the globe. In December 2019, the European Parliament and the Council of the European Union agreed to set a legally binding drinking water limit for PFAS detected in drinking water [18].

Cost of treatment and technologies for removing PFAS from drinking water

Given the chemical properties of PFAS, such as high mobility and solubility, conventional drinking water treatment processes such as coagulation, sedimentation and filtration are not effective for removing these compounds from water [15, 19]. Treatment technologies for PFAS include granular activated carbon, ion exchange and high-pressure membrane filtration. Selection of water treatment options to remove PFAS requires a case by case evaluation for each water system. Initial concentration of PFAS, type of PFAS (long or short chain, carboxylate or sulfonate functional groups), co-occurring contaminants, and water chemistry such as pH and salinity all influence the effectiveness of PFAS removal [19].

Examples of capital and operating costs for PFAS treatments systems are listed in **Table 2**, with a focus on communities studied by Evans et al. [14]. Costs of treatment vary greatly depending on the type of technology selected and system size, or the volume of water treated on a daily basis. All costs are listed in USD, as published at the time of the project, and have not been adjusted to a single year value of USD. As demonstrated in Table 2, reverse osmosis treatment has the highest cost. While this technology is not widely applied at the moment, two new reverse osmosis plants are constructed in the Alabama and North Carolina, with expected completion in 2020 to 2022. GAC filters, due to lower cost, are typically the first option considered for PFAS removal. Overall, cost considerations are the major factor limiting the installation of PFAS removal technologies, and even water testing for PFAS. Residents in communities with PFAS-contaminated water have called for the cost of the water treatment to be borne by the companies and other entities that caused this pollution.

Granular activated carbon (GAC) treatment

GAC filtration is common both at water treatment plants and for point-of-use filters in homes and businesses. Factors impacting the efficacy of this type of treatment include the type of carbon, contact time and the amount of natural organic carbon in the water which can reduce the ability of GAC to remove PFAS. Studies have shown that

perfluorinated sulfonates are more readily adsorbed by activated carbon than perfluoroalkyl acids; longer-chain PFAS are more readily adsorbed than shorter-chain compounds [19-21]. For short chain PFAS, longer GAC media contact time and more frequent media changes are required. Additionally, the source of GAC can influence the effectiveness of PFAS removal. GAC prepared from bituminous coal has been more effective compared to GAC prepared from other materials, such as coconut shell-based GAC [22, 23]. The removal efficiency also depends on how long the GAC is in service before regeneration or replacement [20, 21], and timely replacement of GAC filters is essential. Once the activated carbon has absorbed contaminants to its maximum capacity, contaminant "break-through" is observed, and contaminants absorbed previously can be released to the flow through water [19].

In a case study, researchers from Calgon Carbon Corporation, a company that specializes in GAC production, found that coal-based re-agglomerated GAC achieved cost-effective PFAS removal with an operational cost of \$0.12/1000 gallons treated and an estimated construction cost of \$275,000 per million gallons per day of treated water [22]. Operational costs dropped to \$0.08/1000 gallons if carbon is regenerated [22]. In a simulated small-scale column test study, GAC could reduce the concentrations of both long-chain and short-chain PFAS and that treatment could be effective for one to two years before the required replacement of carbon material [21].

All PFAS removal treatments have to face the difficult challenge of disposal for the PFAS residuals, such as used GAC media, which must be regenerated or transferred to a landfill or an incinerator for combustion at temperatures greater than 1,000 °C. Thermal regeneration involves high temperature heating similar to incineration, which can allow GAC media to be reused. Yet, during this process, fluorinated chemicals are released into the air [24]. Both landfill disposal and thermal regeneration/incineration can cause adverse impacts on human health and the environment. Incomplete incineration of PFAS-containing materials cycles fluorinated chemicals back into the atmosphere, and, with rain, back into the water. More research, community involvement and government action are needed to ensure that the disposal practices for PFAS residuals do not simply move the environmental contamination problem from one media to another.

Reverse osmosis (RO) treatment

Membrane separation is the most effective method for removal of all types of longer- and shorter-chain PFAS tested, including PFOS, PFOA, PFBS, PFHxS, PFHxA and PFNA, as well as the most expensive one [19, 20, 23, 25]. Reverse osmosis generally achieves better contaminant removal than nanofiltration. Membrane separation technology can be combined with GAC for higher removal rates and longer lifetime for the membranes.

Compared to other PFAS removal options, membrane filtration requires significantly more energy [20]. Of note, reverse-osmosis treated water is more corrosive for pipes and plumbing fixtures. Additionally, since drinking ion-free water can lead to nutritional deficiencies, reverse osmosis-treated water needs to be remineralized. Finally, similar to GAC, disposal solutions must be found for the membrane "reject water", which concentrates the source water PFAS. Discharge of the "reject water" back into the environment is equivalent to sending the PFAS pollution downstream where it would contaminate other communities' water sources.

Anion exchange

Anion exchange treatment, a newer technology for removal of PFAS, can be very effective. Similar to GAC, anion exchange is more successful at removing longer chain PFAS compared to shorter chain PFAS [19, 26, 27]. The initial cost of installing anion exchange treatment can be greater than GAC; on the other hand, anion exchange materials require less frequent replacement compared to GAC. Background water chemistry influences the effectiveness of anion exchange technology for PFAS removal, due to the potential presence of competing anions. Single-use resin can be effective, and more research may be needed on regenerable resin [19, 20, 26]. The spent resin needs to be incinerated or transferred to a landfill, similar to the GAC media. Anion exchange can also be used in tandem with GAC, and combination of anion exchange with nanofiltration provides superior removal efficiency compared to GAC or anion exchange alone [28, 29].

Addressing the sources of PFAS contamination

It is an unenviable prospect for a community to have to remove PFAS from its drinking water in perpetuity. In order to protect water supplies from PFAS, it is essential to identify and prevent or remedy the existing sources of PFAS contamination. Contamination of PFAS in groundwater near military bases and airports has been reported from numerous countries and regions. Positive steps have already been taken in this direction, such as the development of new, fluorine-free firefighting foam [30] and the U.S. congressional mandate for a phase-out of PFAS in firefighting foam in 2024.

Municipal wastewater treatment plants, which often receive industrial PFAS wastewater, can transform larger molecular weight PFAS into smaller, more mobile perfluoroalkyl acids. A study in Australia analyzed influent and effluent samples from 19 wastewater treatment plants and reported that the concentrations of one type of PFAS, perfluorocarboxylic acids, were higher in effluent compared to influent, suggesting that larger PFAS precursor molecules are degraded into smaller PFAS compounds during wastewater treatment [10]. PFAS contamination of

wastewater effluent poses a risk for water recycling efforts such as the recharge of aquifers with treated wastewater [31]. In addition to PFAS presence in the wastewater effluent, PFAS also end up in biosolids (sludge) [32, 33] which are applied to the agricultural fields. PFAS contamination in the soil acts as a source of PFAS in food crops [34] and a reservoir for the ongoing PFAS contamination in aquatic ecosystems and aquatic organisms. As more awareness grows about PFAS contamination, companies are starting to produce PFAS-free consumer products. Many apparel companies have moved to eliminate PFAS in their textile treatments [35]. IKEA, a global furniture retailer, reported a complete phaseout of PFAS in products in 2016 [36]. Major home retailers in the U.S. have announced that carpets and rugs they sell will no longer contain PFAS [37, 38]. Such steps hold the promise of decreasing future PFAS releases.

Changing landscape of PFAS contaminants

Since the early 2000s, the regulatory scrutiny has focused on two members of the PFAS family, PFOA and PFOS. Yet, restrictions on PFOA and PFOS production and use have not solved the PFAS contamination crisis. New PFAS chemicals, sometimes called “alternative” or “substitute” PFAS, are continuously developed. Hexafluoropropylene oxide dimer acid (GenX), introduced to replace PFOA in industrial manufacturing, has become a new, persistent contaminant emitted from fluorochemical industrial facilities [39]. A review article published in 2019 noted two aspects of the changing landscape of PFAS contamination: PFAS alternatives are increasingly detected in the environment and in human tissues; and these PFAS chemicals exhibit toxic effects similar to the “legacy” PFOA and PFOS chemicals [40]. According to this study, PFBA, PFBS, PFPeA, PFHxA, and PFHpA were the five most common short-chain PFAS in the environment [40]; and these chemicals were detected in drinking water in the U.S. [14].

As documented in toxicological reviews published by the U.S. EPA, short-chain PFAS compounds GenX and PFBS can harm the kidney and the developing fetus; GenX also affects the immune system and has carcinogenic activity, while PFBS impacts the thyroid and reproductive organs [41, 42]. In animal studies, GenX also caused changes in the thyroid hormones [43]. The U.S. National Toxicology Program recently concluded that short-chain PFAS (PFHxA, PFBA and PFBS) produce toxicological effects similar to long-chain PFOA, PFOA, PFNA, PFDA, and PFHxS, such as liver toxicity and changes in thyroid function [44, 45].

In animal studies, long chain PFAS affect fetal growth and development and cause birth weight reductions, an effect also observed in human epidemiological studies [46]. Short chain PFAS compounds PFBS, GenX, and PFHxA can cause developmental delays and reduced pup weight in laboratory animals [41, 42, 47]. Two other toxicological

effects associated with PFAS should be highlighted: metabolic disruption and immune toxicity. Excess growth and weight gain have emerged as a health effect associated with both long and short chain PFAS in human studies [48, 49]. Weight gain/obesity and metabolic disruption associated with PFAS exposure are likely related to PFAS effects on the growth and differentiation of fat cells [50]. Even at low doses, PFOA, PFOS, PFNA, and PFHxS can cause immune suppression in laboratory animals and in people [51, 52]. Data on potential immunotoxicity of short-chain PFAS are sparse, although in vitro studies indicate that PFBS can suppress cytokine secretion by immune cells similar to the long chain PFAS, PFOA, PFOS, PFDA, and PFOSA [53]. Overall, while short chain PFAS can have a shorter residence time in the body, their toxic potency may be similar to long-chain PFAS, especially when they are present as mixtures of multiple PFAS compounds [54, 55].

Conclusions

In closing, a growing list of research studies documents the threat that PFAS pollution poses to drinking water safety. Communities whose water supplies are impacted by industrial waste discharges from fluorochemical production and PFAS-based firefighting foam are struggling with the economic and health costs of contaminated drinking water. To address this crisis, it is essential to stop industrial discharges and to implement a transition to non-PFAS-based firefighting technologies. To this day, severe challenges remain around PFAS disposal and the presence of PFAS in wastewater effluent from municipal wastewater treatment plants. In the long term, preventing ongoing PFAS releases is needed to protect public health.

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