Letter

Assessing the Effectiveness of Point-of-Use Residential Drinking Water Filters for Perfluoroalkyl Substances (PFASs)

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ABSTRACT: Per- and polyfluoroalkyl substances (PFASs) have come under increased scrutiny due to concerns about their potential toxicity and prevalence in the environment, particularly drinking water. PFASs are difficult to remove in full-scale water treatment systems because of their physicochemical properties. Here we evaluated the effectiveness of point-of-use (POU) and point-of-entry (POE) residential drinking water filters in removing a suite of three perfluoroalkyl sulfonic acids, seven perfluoroalkyl carboxylic acids, and six per- and polyfluoroalkyl ether acids in homes in central ($n = 61$) and southeastern ($n = 12$) North Carolina. POU systems included countertop and pitcher filters, faucet-mounted filters, activated carbon block refrigerator filters, activated carbon block under-sink filters, under-sink dual-stage filters, and under-sink reverse osmosis filters. All under-sink dual-stage and reverse osmosis filters tested showed near complete removal for all PFASs evaluated. In contrast, all other filters containing activated carbon exhibited variable PFAS removal. In these filters, PFAS removal efficiency was dependent on chain length, with long-chain PFASs ($\sim$60−70% removal) being more efficiently removed than short-chain PFASs ($\sim$40% removal). A few whole-house activated carbon POE systems ($n = 8$) were also evaluated; however, results were variable, and in some cases (four of eight systems), increased PFAS levels were observed in the filtered water.
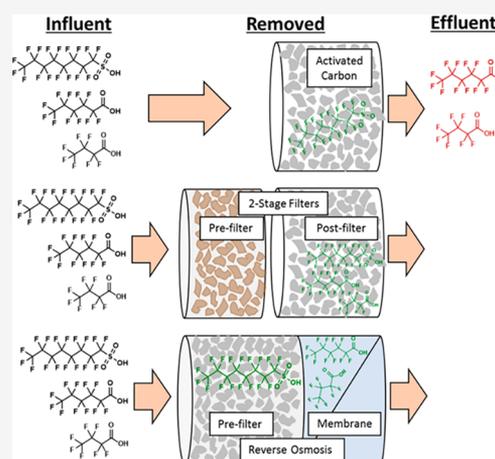
INTRODUCTION

Per- and polyfluoroalkyl substances (PFASs) include several thousand chemicals that have been used in a variety of applications for more than 60 years, including stain- and water-repellent technologies and firefighting foams.[1−11] As a result of widespread usage, they have been detected in most environmental matrices, including soil, groundwater, surface water, ambient air, and house dust.[3,5,7,9,10,12−14] They have also been detected nearly ubiquitously in human serum.[15−19]

Due to the widespread environmental presence of PFASs, their usage has come under increased scrutiny. Within the past decade, long-chain per- and polyfluoroalkyl acids (PFAAs) have been phased out of production and replaced with short-chain PFAAs and per- and polyfluoroalkyl ether acids (PFEAs) such as GenX, ADONA, and F-53B, which are becoming more prevalent in the environment.[4,8,10,20−29] In addition, recent advances in high-resolution mass spectrometry led to the discovery that fluorochemical producers release PFEAs as manufacturing byproducts.[26,30] Long-chain PFAAs are defined as perfluoroalkyl carboxylic acids (PFCAs) with eight or more carbons and perfluoroalkyl sulfonic acids (PFSAs) with six or more carbons.[2]

PFAAs, particularly long-chain PFAAs, have a wide range of potential toxicological effects.[31] They have been associated with various cancers and non-Hodgkin's lymphoma,[32] linked to thyroid disorders,[17,33,34] and shown to have immunotoxic effects,[35] among other outcomes. Most toxicological studies focus on long-chain PFAAs, specifically PFOA and PFOS, and less is known about the toxicological effects of short-chain PFAAs, such as perfluorohexanoic acid (PFHxA), or PFEAs such as GenX. Current research shows that when compared to their long-chain counterparts, some short-chain PFAAs appear to be as persistent in the environment[8,20−23] and less bioaccumulative in animals and humans.[36−38] While people can be exposed to PFASs in a variety of ways, current research suggests that an important exposure pathway for the general population is through ingestion of contaminated drinking water.[39−42]

Regulatory agencies have proposed guidelines and standards in recognition of the potential negative health effects associated with PFAS exposure through drinking water. The U.S. Environmental Protection Agency (U.S. EPA) has set a lifetime health advisory level of 70 parts per trillion (ppt or nanograms per liter) for the combined levels of PFOA and PFOS in drinking water.[43−45] Several U.S. states are moving toward more stringent regulations.[46,47] For example, in late 2018, the New Jersey Department of Environmental Protection (NJDEP) adopted an MCL of 13 ppt for PFNA, the first enforceable regulation of its kind.[47]

Despite increased awareness and proposed regulations, PFAAs are repeatedly detected at elevated levels in treated drinking water.[9,25,48−55] PFASs are not always removed in traditional full-scale water treatment systems due to their physicochemical properties.[48−50,52−54] Treatment technologies that can effectively remove PFASs in drinking water treatment plants include granular activated carbon (GAC) adsorption, anion exchange, and high-pressure membrane filtration, such as nanofiltration and reverse osmosis. While GAC and anion exchange resin have varied success with smaller PFASs, reverse osmosis systems can consistently remove PFASs to below detection limits.[48,50,52−56] Though a few studies have examined POU systems in a laboratory setting,[57,58] no studies to date have examined removal efficiencies in point-of-use (POU) filters in a residential setting.

This study was designed to assess the removal effectiveness of POU filters, including pitcher, refrigerator, under-sink, faucet-mounted, dual-stage, and reverse osmosis systems, as well as POE systems (Table S9), for three PFSAs, seven PFCAs, and six PFEAs (Table S1). The PFSA and PFCA assessment was focused on central North Carolina (NC) communities. A majority of the PFEA assessment was specifically targeted in southeastern NC near the coastal town of Wilmington, NC, and the surrounding area because of elevated levels present in the water supply.[25]

MATERIALS AND METHODS

Central NC samples (n = 63 water samples from 61 homes) were collected between May 2018 and March 2019 from a convenience sample residing in Chatham, Durham, Orange, and Wake counties. A convenience sample of North Carolina homeowners was recruited for the collection of water samples using flyers (posted throughout the community, on social media, and on local listservs) and word of mouth. Homeowners provided information about their water utility and information about their water treatment systems (e.g., purchased date, filter replacement, etc.). Detailed sampling and laboratory procedures, as well as QA/QC results, can be found in the Supporting Information. The southeastern NC samples (n = 26 water samples from 12 homes) were collected from a convenience sample residing in New Hanover and Brunswick counties in southeastern NC between June 30, 2017, and December 19, 2017. Detailed methods used for the southeastern NC samples are available in the Supporting Information and have been published elsewhere.[30] Two source waters in central North Carolina that provide water for Pittsboro and Cary, NC, were also sampled between August and November 2019. The results from this analysis are available in the Supporting Information.

RESULTS AND DISCUSSION

In total, we collected samples from 73 homes, with 87 unfiltered/tap and 89 filtered water samples (Tables S2 and S3). Some homes were visited more than once (particularly in the southeastern NC cohort), and several homes had more than one filter that was tested, resulting in more filtered samples that were tested compared to unfiltered samples.

Prefilter Tap Water Characteristics. Raw unfiltered tap water was assessed between May 2018 and March 2019 for PFSAs and PFCAs within communities served by five water utilities in central North Carolina, specifically the Durham County Utilities Division, Orange Water and Sewer Authority (OWASA; serving Chapel Hill and Carrboro), the City of Cary, the City of Raleigh, and the Town of Pittsboro. Private well water samples (n = 4) were also collected for comparison. The median $\sum$(PFCA+PFSA) concentration across all water utilities was 43 ng L^{-1} and ranged from 6 to 759 ng L^{-1}. The utilities arranged from highest to lowest median $\sum$(PFCA+PFSA) concentration are Pittsboro (56 ng L^{-1}; range of 31−759), Cary (53 ng L^{-1}; range of 21−91), OWASA (49 ng L^{-1}; range of 6−66), Raleigh (20 ng L^{-1}; range of 17−22), and Durham (14 ng L^{-1}; range of 10−32) (Figure S1). All well water samples had a $\sum$(PFCA+PFSA) concentration below 15 ng L^{-1} (median of 8 ng L^{-1}; range of 1−16). Although GenX and other PFEAs have been found at relatively high levels in the Cape Fear River basin (southeastern NC),[25] they were detected at only low levels in our central NC water sample with a median concentration of 0.7 ng L^{-1} (range of 0.2−1.4 ng L^{-1}) across all central NC utilities.

In June 2017, samples collected in southeastern NC had a $\sum$(PFCA+PFSA) concentration of 123 ± 1 ng L^{-1} and a GenX concentration of 202 ± 32 ng L^{-1}. In addition, high levels of other PFEAs were present, for which we did not have analytical standards at the time this study was conducted. A recent analysis of an archived Cape Fear River sample from June 2015 yielded a summed PFEA concentration of ~100000 ng L^{-1}.[59] Prior to subsequent sample collection in southeastern NC, the upstream fluorochemical manufacturer took steps to decrease the discharge of GenX and other PFEAs. In August 2017, the $\sum$(PFCA+PFSA) concentration was 105 ± 35 ng L^{-1} and the GenX concentration had decreased to 55 ± 12 ng L^{-1}. In December 2017, the $\sum$(PFCA+PFSA) concentration was 165 ± 20 ng L^{-1} and the GenX concentration was 45 ± 17 ng L^{-1}. Variability in the $\sum$(PFCA+PFSA) concentrations over the June to December 2017 time period can be explained by variability in streamflow as well as upstream PFCA and PFSA inputs.

Effectiveness of Point-of-Use (POU) and Point-of-Entry (POE) Filters. Of the 89 total filtered water samples, 76 were POU systems and 13 POE systems. Of the 76 POU systems, 18 were reverse osmosis filters, seven were dual-stage filters, 10 were under-sink activated carbon block filters, two were faucet-mounted, one was a countertop unit, 13 were pitcher filters, and 25 were refrigerator filters. Of the 13 POE systems, eight were GAC-based and five had both GAC and ion exchange technology incorporated. These filters ranged in age from <1 month to >5 years, with an average reported age of 7.5 months.

We examined several filter characteristics that may explain the variability in removal efficiency across all filters and PFASs, but we did not find any statistically relevant correlations between removal efficiency and brand, source water matrix/

Table 1. Average Percent Removal for PFAAs and PFEAs by Filter Class[a]

		Central NC								
		counter filter (n = 0)	faucet filter (n = 2)	pitcher filter (n = 13)	fridge filter (n = 22)	single-stage under-sink filter (n = 5)	whole-house, GAC (n = 6)	whole-house, GAC/CIX (n = 0)	two-stage filter (n = 4)	reverse osmosis (n = 11)
PFSA	PFBS	na	94%	65%	29%	>84%	18%	na	>92%	94%
	PFHxS	na	88%	54%	65%	>84%	32%	na	>95%	>96%
	PFOS	na	99%	71%	61%	>99%	67%	na	99%	100%
PFCA	PFBA	na	63%	36%	45%	15%	−34%	na	98%	>98%
	PFPA	na	67%	42%	35%	52%	−85%	na	>99%	>99%
	PFHxA	na	79%	43%	59%	53%	−63%	na	>97%	98%
	PFHpA	na	75%	43%	65%	52%	−37%	na	>97%	98%
	PFOA	na	84%	67%	71%	56%	19%	na	>99%	>92%
	PFNA	na	92%	>54%	72%	45%	28%	na	>99%	>88%
	PFDA	na	99%	>57%	57%	64%	44%	na	>99%	>93%
PFEA	GenX	na	63%	46%	56%	51%	21%	na	>99%	>99%
		Southeastern NC								
		counter filter (n = 1)	faucet filter (n = 0)	pitcher filter (n = 0)	fridge filter (n = 3)	single-stage under-sink filter (n = 5)	whole-house, GAC (n = 2)	whole-house, GAC/CIX (n = 5)	two-stage filter (n = 3)	reverse osmosis (n = 7)
PFEA[b]	GenX	−146%	na	na	>78%	>74%	55%	−7%	>74%	>75%
	PFMOAA	−498%	na	na	92%	91%	40%	−106%	97%	97%
	PFO2HxA	−284%	na	na	98%	94%	54%	58%	>99%	97%
	PFO3OA	11%	na	na	>99%	97%	64%	60%	99%	>99%
	PFO4DA	70%	na	na	99%	99%	63%	69%	99%	99%
	Nafion BP2	55%	na	na	97%	>99%	52%	1%	98%	99%

[a]All measurements displaying negative percent removal values experienced an increase in PFAS concentration after filtration. [b]Percent recoveries calculated for the southeastern NC PFEA measurements were determined with peak area counts normalized to internal standards.

loading, or age of the filter. For this reason and because of our limited sample size, much of the analysis and discussion surrounding the POU and POE filters is done with percent removal as the primary metric. Raw concentration values for all samples collected are available in Table S9.

Upon comparison of pre- and postfilter $\sum$(PFCA+PFSA) concentrations across all central NC samples, there was significant removal by POU/POE filters ($p < 0.001$; Wilcoxon-Rank sum test). To assess the effectiveness of each individual filter, a Wilcoxon-Rank sum test was conducted on the pre- and postfilter levels across compounds for all samples (Table S7). On the basis of this analysis, 79% of samples (50 of 63) showed significant ($p < 0.05$) removal, while 21% (13 of 63) did not show significant ($p > 0.05$) removal.

Upon examination of the removal effectiveness by filter type, all reverse osmosis and dual-stage filters showed significant removal ($p < 0.05$) for PFCAs and PFSAs, reducing levels at a $\geq 90\%$ removal efficiency (Table 1). Similarly, all reverse osmosis and dual-stage filters examined for PFEAs reduced target compound levels at a $\geq 97\%$ removal efficiency, with the exception of GenX (Table 1). GenX was removed at >74% and >75% efficiency in dual-stage and reverse osmosis systems, respectively. GenX was below MDL levels in the filtered samples, and given the low levels in the unfiltered samples and the MDL, we could not determine removal percentages in excess of ~75%. This clearly demonstrates the effectiveness of membrane and dual-stage filters for removing both long- and short-chain PFAAs, as well as novel PFEAs. It is unsurprising that reverse osmosis systems were effective, given that these systems often include a prefilter (sometimes activated carbon-based) followed by a semipermeable membrane that removes all but the smallest contaminants. It is unclear why the dual-stage filters were effective; it is hard to reach any generalizable conclusion given our small sampling of dual-stage filters.

Apart from the dual-stage filters, activated carbon POU filters (including countertop, faucet, pitcher, fridge, and single-stage under-sink filters) demonstrated much greater variability across our study with 73% of all activated carbon filters showing significant removal (Wilcoxon-Rank sum test) for PFSAs and PFCAs (Table S7). On average, single-stage under-sink filters ($n = 5$) removed a majority of PFSAs (>84% removal) but removed only half of PFCAs. Similar to the case for the PFSAs, the under-sink filters in southeastern NC ($n = 7$) removed a majority of PFEAs (>90% removal). The poor performance of the under-sink filters at removing PFCAs is particularly surprising given the favorable performance of the dual-stage filters previously discussed. Further work is needed to understand the importance of the sediment filter and the size of the ACB filter on PFAS removal efficiency.

The two filter categories with the greatest number of samples analyzed were pitcher filters and refrigerator filters, and both performed with comparable efficiencies. For the pitcher filters, 85% (11 of 13) showed significant removal for PFCAs and PFSAs while 68% (15 of 22) of refrigerator filters showed significant removal for PFCAs and PFSAs. Both filter types provided ~50% removal for all target PFCAs and PFSAs measured, with increased removal efficiency for long-chain PFAAs (61% for pitcher filters and 65% for refrigerator filters) compared to short-chain PFAAs (46% for pitcher filters and 47% for refrigerator filters). The data for pitcher filters may suggest superior removal for PFSAs compared to PFCAs, but statistical significance was not reached. Refrigerator filters in southeastern NC removed all six target PFEAs more efficiently (>92% removal) than refrigerator filters in central NC removed PFAAs (approximately 50% removal). It is important to note that refrigerator filters are offered in a range of sizes and contain activated carbon quantities that differ in mass by a factor of at least 3.[60]

We examined two faucet-mounted filters in central NC for removal of PFSAs and PFCAs, both of which were relatively new (<1 month old), and the filters displayed favorable performance (>75% removal, on average). Similarly, we tested only one countertop unit in southeastern NC that was ∼2 years old and performed poorly for PFEA removal. With the current data on these two filter types, we cannot reach any generalizable conclusions.

The large POE whole-house granular activated carbon filters analyzed in this study were found to have mixed results. Half of the filters were found to have significant removal of PFAS, and half had nonsignificant removal (Table S8). Four of the six whole-house carbon filter systems we analyzed for PFCAs and PFSAs experienced an increase in $\sum$(PFCA+PFSA) concentration after filtration, suggesting saturated media in these larger activated carbon systems may be desorbing PFASs into the home's water. In southeastern NC, we examined two whole-house carbon filter systems for PFEAs. One of the systems had favorable removal (>80% removal on average), while the other had poor overall removal (<30% on average). In addition to the two whole-house activated carbon filters, we examined five whole-house systems with ion exchange technology. PFAS removal in these systems was largely inconsistent and unpredictable. Some showed favorable removal for some PFEAs while simultaneously exhibiting increases for other target PFEAs. While our sample size is small, the postulation that these systems may be desorbing into a home's water supply has been observed in other studies examining full-scale treatment systems.[48,50,52,54,61] The ability to predict whole-house system performance is further complicated by the fact that these systems often contain proprietary media apart from activated carbon, and it is therefore difficult to make any overarching claims about the effectiveness of POE systems.

General Observations. Despite the variability, we found that on average PFAAs with greater chain lengths were more efficiently removed by activated carbon filters. We also found that on average PFSAs were removed better than their PFCA counterparts of equal chain length (Figure 1 and Table 1). PFSAs are more efficiently removed by activated carbon because PFSAs are more hydrophobic than their PFCA counterparts of equal chain length.[62] Although there is large variability within our study, we found a significant ($p < 0.05$) positive correlation between the average percent removal by
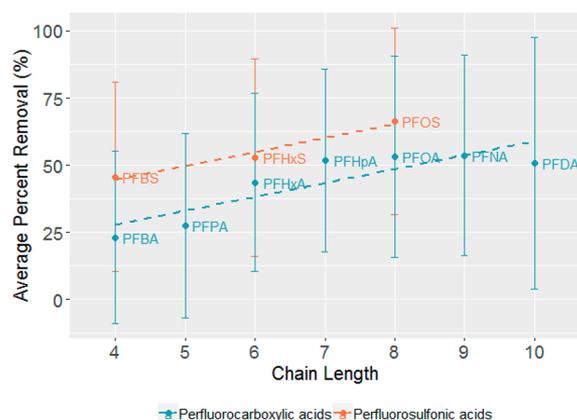
activated carbon filters and chain length for PFCAs. We found a similar trend for the PFSAs, though statistical significance was not reached (Figure 1). This relationship between chain length and removal effectiveness is consistent with what others have found for full-scale water treatment systems.[25,52,55] A similar trend was seen for the PFEAs based on the size of the molecule. The smallest PFEA molecule we analyzed, PFMOAA (MW = 180), was generally the least efficiently removed across filter classes, and removal efficiency improved as the molecule size increased (Table 1).

These results suggest that short-chain PFAAs are not reliably removed by activated carbon POU filters, with the exception of dual-stage filters. In our study, short-chain PFAAs had an average percent removal of 41% by activated carbon-based filter systems. This is particularly important as replacement PFAAs (short-chain PFAAs and novel PFEAs) were dominant in several of the regions we studied, accounting for a majority of the total PFAA source water concentrations. While short-chain replacement PFAAs are likely less bioaccumulative in the environment than their long-chain counterparts, they are still persistent and are becoming more prevalent in the environment. In fact, there is already some evidence suggesting that this shift has occurred.[63−67] For example, Glynn et al. conducted a temporal trend analysis of serum PFAA levels in a Swedish cohort and found that serum levels of replacement PFAAs like PFBA and PFHxS increased ∼10% per year from 1996 to 2010 while levels of legacy PFAAs like PFOA and PFOS decreased between 1996 and 2010.[64] Despite their widespread usage, much is still unknown about the environmental and toxicological effects of replacement PFAAs and more research is needed.

While AC-POU systems fail to remove many short-chain PFAAs, they do reduce long-chain PFAA concentrations by 60−70% on average. Previous studies have found a statistically significant reduction in PFAA serum levels for people who report using POU filters.[51,68] This suggests that activated carbon-based filters significantly reduce exposures despite varied performance efficiencies.

The goal of exposure reduction should be removal of PFAS contamination from the source water, prior to drinking water treatment; however, in some cases, identifying the source of PFAS can be challenging. Therefore, residents may need to consider POU systems to reduce their personal exposures in the home. We demonstrated that residential activated carbon POU/POE systems have variable performance. While under-sink reverse osmosis systems appear to be an ideal option for removing PFAS, they have a high capital cost. Ultimately, an activated carbon filter should provide some removal of PFAS from drinking water (particularly the long-chain PFAAs) and have a lower capital cost.

■ ASSOCIATED CONTENT

ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.0c00004.

Target PFAS information, method information, quality assurance data, tabulated statistics, and additional figures (PDF)

Figure 1. Average percent removal compared to chain length of PFAAs for activated carbon-based point-of-use filters.

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Notes

The authors declare no competing financial interest.

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