

EMERGING CONTAMINANT ARTICLE

Longitudinal assessment of point-of-use carbon filters for removal of per- and polyfluoroalkyl substances from private well water

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Funding information

North Carolina Policy Collaboratory, Grant/Award Number: N/A

Associate Editor: Lauren Weinrich

Abstract

Eighteen private well users in North Carolina were recruited to test the performance of under-sink, activated carbon block water filters to remove per- and polyfluoroalkyl substances (PFAS). Monthly sampling was conducted for 8 months. Filters were certified for removal of perfluorooctanoic acid and perfluorooctanesulfonic acid under NSF International certification P473, but not for additional short-chain perfluoroalkyl acids or perfluoroalkyl ether acids (PFEAs) evaluated in this study. Out of 47 targeted analytes, 17 PFAS were detected in filter influent samples (influent \sum PFAS 4.7–131 ng/L). Mixed-effects Tobit regression models showed that the filters effectively removed 97%–99% of all influent PFAS, including short-chain PFEAs, for the entire manufacturer-recommended lifetime of the device. The prevalence of PFAS above the minimum reporting limits was reduced by 99.5%, and the prevalence of any PFAS above the method detection limits was reduced by 92%. The results provide increased confidence in NSF P473-certified filters for the removal of PFAS from private well water.

KEYWORDS

activated carbon, PFAS, point-of-use water treatment, private wells

1 | INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS)—including perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkyl

sulfonic acids (PFSAs) (together referred to as perfluoroalkyl acids (PFAAs)), and replacement perfluoroalkyl ether acids (PFEAs)—have generated concern in recent decades for their widespread environmental

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occurrence (Hu et al., 2016; Nakayama et al., 2007; Pan et al., 2018; Sun et al., 2016) and adverse human health effects. Elevated exposure to two legacy PFAS that have been in production since the 1940s, perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS), has been associated with liver damage, increased risk of thyroid disease, increased cholesterol, and possible carcinogenic effects (ATSDR, 2021; Grandjean & Clapp, 2015; Steenland et al., 2010).

As a result, long-chain PFAAs like PFOA and PFOS have been phased out of production in recent decades and the U.S. Environmental Protection Agency (USEPA) has set a lifetime Health Advisory Level (HAL) of 70 ng/L in drinking water for PFOA and PFOS combined (USEPA, 2016a, 2016b). Alternative PFEAs have been developed to replace legacy PFAAs, such as the ammonium salt of hexafluoropropylene oxide dimer acid (HFPO-DA, also known by its trade name GenX). Research is ongoing to determine the toxicity of these compounds to establish drinking water regulations in the United States (Conley et al., 2019; Guelfo et al., 2018; USEPA, 2021; Wang et al., 2017).

Drinking water may contribute a significant portion of overall PFAS exposure, particularly for those relying on private drinking water wells near contaminated sites, as documented in areas of Ohio and West Virginia (Hoffman et al., 2010), Minnesota (Xiao et al., 2015), New Hampshire (Daly et al., 2018), Colorado (Barton et al., 2020; McDonough et al., 2021; Starling et al., 2019), North Carolina (Roostaei et al., 2021), and elsewhere. Private wells that are distant from hazardous waste sites and industrial point sources may still contain PFAS, however, from the cumulative impacts of background sources such as septic systems (Schneider et al., 2014, 2016), legacy municipal landfills (Hepburn et al., 2019), historical applications of aqueous film forming foam (AFFF) for fire suppression (Weber et al., 2017), and/or rural applications of biosolids (Lindstrom et al., 2011). Given the prevalence and toxicity of PFAS, private well users need evidence-based interventions to limit drinking water exposures.

Household and point-of-use (POU) water filters have been implemented as a possible intervention for well users impacted by PFAS-contaminated groundwater on both an ad hoc and legal compliance basis. Examples include the purchase of a range of water filter types by private well users in an area of groundwater contamination in Colorado (Patterson et al., 2019) and the distribution of whole-house granular activated carbon (GAC) and POU reverse osmosis (RO) filters to well owners impacted by the spread of PFAS surrounding a fluoropolymer manufacturing facility in North Carolina (NCDEQ, 2019; North Carolina General Court of Justice, 2019). To aid consumer decision-making regarding these devices, NSF International (NSF)

Article Impact Statement

Activated carbon point-of-use water filters may be used to help protect private well users from per- and polyfluoroalkyl substances exposures.

released protocol P473 to test and certify household water treatment products for the removal of PFOA and PFOS to below the USEPA HAL. Other entities such as *Consumer Reports* have also begun to provide independent testing of POU filters that may be effective for PFAS (Santanachote & Bergman, 2021) and even make recommendations for maximum limits of PFAS in drinking water (Felton, 2021).

Limited information exists for individual well users and policymakers to make informed decisions regarding the long-term effectiveness of these devices, however. To date, only three other peer-reviewed studies have assessed the effectiveness of residential POU filters for PFAS, and none have evaluated their performance on private well water. First, Anumol et al. (2015) demonstrated that both pour-through and refrigerator POU filters using activated carbon have some capacity to remove PFOA and PFOS. Performance of pour-through filters comprised of loose GAC media was highly variable, but refrigerator filters using solid activated carbon blocks (ACB) removed >97% of PFOA and PFOS for the entire manufacturer-stated treatable volume from a municipally treated groundwater. A second POU treatability study was conducted by Patterson et al. (2019) using commercial RO units and GAC. RO units and GAC column tests both demonstrated effective removal of six legacy PFAAs associated with AFFF with influent challenge concentrations of over 5600 ng/L total PFAS. Lastly, Herkert et al. (2020) tested household performance of POU devices, including ACB filters, in a cross-sectional study of 61 households in North Carolina where the total PFAS concentrations ranged from 6 to 759 ng/L. RO and dual-stage ACB filters were shown to remove >74%–99% of both long- and short-chain compounds, including PFEAs, in municipal tap waters.

While these studies suggest that POU filters may provide an effective solution for private well users concerned about or impacted by PFAS, important knowledge gaps remain. Anumol et al. (2015) and Patterson et al. (2019) tested POU treatment effectiveness over time but did not evaluate the numerous emerging PFEAs and short-chain PFAAs now detected in some drinking waters in the United States (Hopkins et al., 2018; Sun et al., 2016). Additionally, these studies were conducted under laboratory-controlled conditions, including constant flow rate and water quality, which are not representative of

variable household conditions and irregular use patterns. Herkert et al. (2020) tested POU devices under household conditions, but removal performance could only be related to estimates of filter age (i.e., time since installation) rather than precise measurements of cumulative volume treated. Cross-sectional measurements of treatment effectiveness are an incomplete indicator of overall protectiveness as the removal performance at a single time point neither provides information regarding the time-to-failure nor accounts for variations in water use. Lastly, none of the previous studies included private well water, limiting the utility of these studies to provide recommendations to well users as the chemistry of private well water may be different from the test waters used during NSF certification and can vary widely from one location to another and over time. In the absence of this information, state health departments have conducted their own tests using POU treatment devices to inform well users in their state (see MDH, 2017), but in situ testing with well users is needed to inform real-world performance.

To begin to fill these gaps in understanding and to provide the public with improved insight around household water treatment for PFAS, this study aimed to evaluate the performance of under-sink ACB filters installed in 18 homes relying on PFAS-impacted well water. By employing a longitudinal study design, various household-level variables such as volume of water treated, patterns of water use, and influent water quality conditions were related to the filters' performance over time. To date, no other studies have provided a longitudinal analysis of the effectiveness of commercially available POU ACB water filters for PFAS removal under household conditions. These results thus provide vitally important information toward decision-making in communities impacted by PFAS in groundwater across the nation.

2 | METHODS

2.1 | Study area and participant recruitment

Eighteen households were recruited to participate from three different communities (A, B, and C) with groundwater quality concerns in Orange County and Robeson County, North Carolina (Figure S1). Study recruitment procedures and demographic characteristics of each area have been described in detail previously (Mulhern et al., 2021; Mulhern et al., 2022; Mulhern & MacDonald Gibson, 2020). Briefly, 11 households were recruited in Robeson County approximately five miles west of a fluorochemical manufacturing facility (cluster C) that had been identified by the Robeson County Health

Department as being impacted by the spread of HFPO-DA from the facility. Under a legal consent order, the company is required to provide replacement water supplies (either bottled water or up to three under-sink RO systems) to households on private wells surrounding the facility if any of 12 specified PFAS (including HFPO-DA) exceeds 10 ng/L or if the sum of the 12 PFAS exceeds 70 ng/L in the household's well water (North Carolina General Court of Justice, 2019). Households that had previously had their water tested for HFPO-DA by the local health department but were below the 10 ng/L threshold remained concerned, however, and were identified as potential candidates for this study. Three additional households were recruited from the west side of Robeson County, approximately 15 miles away from the fluorochemical facility, where prior testing of well water for PFAS had not been conducted (cluster B). Lastly, four households were recruited in Orange County near the county landfill (cluster A), which has been shown to have contaminated nearby drinking water wells but previously had not been tested for PFAS (Heaney et al., 2015). Average influent groundwater quality characteristics within each cluster compared to the required influent water quality for certification under NSF P473 are provided in Table 1. Groundwater in clusters B and C was significantly below the certified pH range, typical of the acidic conditions of surficial aquifers in the North Carolina Coastal Plain (DeSimone et al., 2020). This study was approved by the University of North Carolina Institutional Review Board (IRB Study No. 19-1015).

2.2 | POU treatment system

A commercially available POU, single-stage, ACB filter (AO-MF-ADV, A.O. Smith) was selected as the test device. This filter treats the full flow of cold water at the kitchen tap and is widely available at Lowe's hardware stores for \$100. The manufacturer states two measures of expected lifetime based on volume (2967 L) and time of use (6 months). Each replacement cartridge costs \$70, representing a maintenance cost of \$12/month or approximately \$0.02/L according to the volume-based capacity. By comparison, the price of bottled water can range \$0.20–\$2.20/L, or 10–100 times higher (Pieper et al., 2019).

At the time of selection, the AO-MF-ADV was the only full-flow, under-sink ACB filter available to consumers that was certified for PFOA and PFOS removal according to NSF P473.

The ACB cartridge used in the filter is a hollow, extruded column produced from a coconut shell carbon mixed with a proprietary ion-exchange metals scavenger and a binding agent. Water flows from the outside in

TABLE 1 Average influent groundwater quality characteristics among participating households in each geographic cluster (total $n = 18$) compared to the required influent water quality characteristics for PFOA + PFOS removal certification according to NSF P473

	Cluster A ($n = 4$)		Cluster B ($n = 3$)		Cluster C ($n = 11$)		NSF P473
	Mean	SD	Mean	SD	Mean	SD	
pH	7.1	0.4	5.3	1.0	4.4	0.3	7.5 ± 0.5
Conductivity ($\mu\text{S}/\text{cm}$)	350	107	168	135	109	35	100–250 ^a
Temperature ($^{\circ}\text{C}$)	17.2	0.3	19.0	1.1	18.7	1.1	20 ± 2.5
DOC (mg/L)	1.2	0.4	0.8	0.8	0.6	0.9	>1.0
Hardness (mg/L CaCO_3)	110.5	50.3	30.2	8.8	17.9	7.2	-

Abbreviations: DOC, dissolved organic carbon; PFOA, perfluorooctanoic acid; PFOS, perfluorooctane sulfonic acid.

^aConverted range for 200–500 mg/L total dissolved solids.

with an approximate bed volume (BV) of 0.95 L. The mass of carbon within the filter cartridge was estimated to be approximately 475 g (Mulhern & MacDonald Gibson, 2020). A sample of the carbon material used to produce the block was obtained from the manufacturer, and the BET (Brunauer, Emmett, and Teller) surface area and micropore volume (pores <2 nm in width) were measured by N_2 gas adsorption using an MP-1 Autosorb instrument (Quantachrome Instruments, Boynton Beach, FL), which indicated a low overall surface area and high microporosity compared to other commercial activated carbons (Table S1).

Water filters were plumbed in underneath the main kitchen tap (Figure 1) using polyethylene tubing and nylon or polypropylene components with an integrated flow meter (Sea YF-S201 or Gredia GR-301) and data logger (HOBO Onset) to measure precise water use over time. The data logger's memory could record at 10-s

intervals for up to 40 days. Sample ports were installed immediately before and after the filter underneath the sink to avoid any confounding results from distant sampling locations at the well head or the faucet. Check valves were installed after each sample port to prevent any backflow where water pressures were low.

2.3 | Sampling protocol

As described previously (Mulhern & MacDonald Gibson, 2020), study participants were trained to collect samples in 1 L, acid-washed HDPE bottles at the influent and effluent sample ports at approximately monthly intervals for 8 months (October 2019–June 2020). Since the device did not have a performance indicator built in to alert the user once the volume capacity had been reached, the study was continued 2 months beyond the

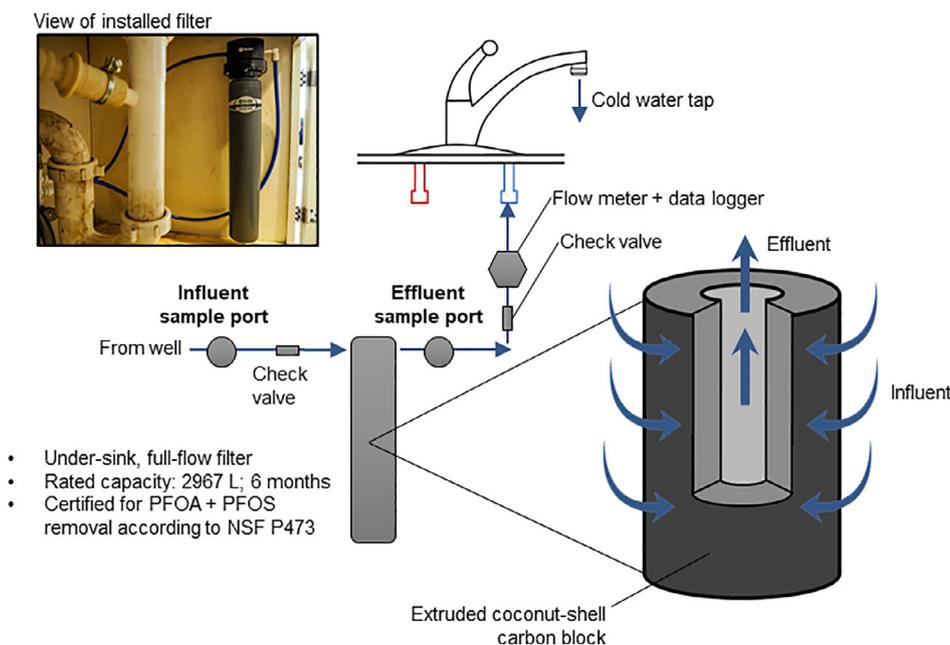


FIGURE 1 Schematic of filter installation underneath the primary kitchen sink in 18 households connected to private wells, adapted from Mulhern and MacDonald Gibson (2020)

recommended 6-month lifetime to evaluate the safety of the device in the event that cartridges are not replaced promptly at the 6-month mark. At the time of each visit, the data logger memory was downloaded and reset by the researchers. The ideal sampling frame of 18 households \times 8 months would have yielded 144 influent/effluent sample pairs (288 samples total), but two sampling months were lost due to Covid-19 restrictions during the study and other challenges (e.g., filters clogging prematurely or study participants not being home) prevented samples from being collected from certain households on some months. Thus, 91 sample pairs (182 samples) were collected in total. Samples were transported to the University of North Carolina (UNC) at Chapel Hill and stored in the dark at room temperature until analysis.

2.4 | Sample storage

Due to laboratory shutdowns caused by Covid-19, samples were stored for seven up to 15 months prior to extraction. Although PFAS losses in water samples have been shown to be negligible for up to 70 days of holding time at room temperature (Sun et al., 2016), changes in PFAS levels may occur in some samples during extended storage times as a result of PFAS transformations (Woudneh et al., 2019). To evaluate the effect of the long holding time, 13 baseline, raw well water samples that were initially collected to determine each household's eligibility to participate in the study in July 2019 were analyzed twice, once after 2 months of holding time (September 2019) and again after approximately 18 months of holding time (January 2020) at room temperature. These samples were not used in the filter evaluation but were only used to determine household eligibility as described elsewhere (Mulhern & MacDonald Gibson, 2020), and for the holding study. The later analyses (January 2020) were conducted according to Method I described below. The early analyses (September 2019) were conducted using a slightly different method using solid-phase extraction (SPE) according to USEPA protocol D-EMMD-PHCB-043-SOP-03 (Tian & Sun, 2019), which was used during early method development but not again for the later analyses. The results of this holding study showed that slight losses in certain analytes were observed after 18 months, including PFOA, PFOS, and HFPO-DA (Figure S2). However, average losses were only 2.5, 1.3, and 1.8 ng/L for these three analytes, respectively. Average losses for all other analytes were less than 2 ng/L. Thus, while certain PFAS transformations may have occurred during the long storage time, they were not considered to significantly affect the filter assessment.

2.5 | Analytical procedures

Two different analytical methods were used for PFAS analyses. The two methods are described briefly below with additional details provided in the Supporting Information.

2.5.1 | Method I: SPE

The primary method used for all influent and effluent samples was weak anion exchange SPE and liquid chromatography/tandem mass spectrometry (LC-MS/MS) adapted from USEPA Method 533 (Hunt et al., 2020; Rosenblum & Wendelken, 2019). The method includes a standard suite of 24 PFAS (Table S2). Before analysis, any samples with a pH outside the range 6–8 were buffered with 1 g/L HPLC-grade ammonium acetate. Due to the possibility of sorption of PFAS analytes to the bottle surface during the extended sample storage time (Rosenblum & Wendelken, 2019), the sample was transferred into a separate, clean container; the inside of the sample bottle was then rinsed with 10 mL of methanol and the entire sample volume was returned to the original container with the rinsate and mixed well by shaking. A volume of 250 mL was then subsampled and spiked with 50 μ L of isotopically labeled PFAS internal standard mix (Wellington EPA-533ES) and mixed well.

PFAS were extracted using a weak anion exchange, polymeric sorbent (Phenomenex #8B-S038-HCH), eluted with 10 mL of methanol with 2% ammonium hydroxide into 15 mL polypropylene conical centrifuge tubes, and concentrated to dryness under high-purity nitrogen in a heated water bath (60°C). Extracts were reconstituted in 0.5 mL of 80% methanol/reagent water (vol/vol) for a final concentration factor of 500. The isotope performance standards (Wellington EPA-533IS) were added to the reconstituted extracts and vortexed to mix. Extracts were stored at room temperature before analysis for no longer than 28 days according to USEPA 533. A 10 μ L aliquot of each sample was injected onto an Agilent 1290 LC system and the eluting compounds quantified on an Agilent 6470 triple quadrupole MS/MS instrument according to conditions specified in Table S4. A six-point calibration curve, prepared neat in 80% methanol using the USEPA 533 native analyte primary dilution standard (Wellington EPA-533PAR) representing a concentration range in the original unextracted samples of 0.5–100 ng/L, was analyzed as above at the beginning of each sample batch and fit using a linear regression. Quality controls included two continuing calibration checks at the low and high end of the calibration curve of each

batch, extracted laboratory reagent blanks, and replicate laboratory fortified blanks (LFBs) spiked at known concentrations at three levels of the calibration curve (5, 10, and 20 ng/L). The relative standard deviation (RSD) and percent recovery of all LFBs were required to be within $\pm 20\%$ and $\pm 30\%$, respectively, for all method analytes (Tables S5 and S6). All analytes passed both quality control criteria except for 6:2 FTS, which was dropped from the method.

The minimum reporting limit (MRL) was determined for each analyte as the lowest concentration on the calibration curve where the mean recovery of seven fortified replicates could be quantified within $\pm 50\%$ with at least 99% confidence according to USEPA protocols (Munch & Branson, 2004; Rosenblum & Wendelken, 2019). The method detection limit (MDL) was also determined to estimate removals where filter effluent concentrations were $< \text{MRL}$. The MDL was defined for each analyte according to USEPA guidelines as the higher of the following: (1) the standard deviation around seven spiked replicates at the MRL for each analyte multiplied by the one-sided Student's *t*-value at 99% confidence and six degrees of freedom or (2) the mean result of all replicates of the method blanks plus the standard deviation times the one-sided Student's *t*-value at 99% confidence and *n*-1 degrees of freedom (USEPA, 2016c). The resulting MRLs and MDLs ranged 0.5–6.2 ng/L and 0.1–3.6 ng/L, respectively (Table S2).

2.5.2 | Method II: Large volume direct injection

Nine paired influent/effluent samples from the last sampling month (June 2020) from a subset of homes in cluster C were also analyzed for 23 additional PFAS not included in USEPA 533 (Table S3), including 14 emerging PFEAs known to be associated with the local fluorochemical manufacturer (North Carolina General Court of Justice, 2019). The method entailed direct injection of the sample (i.e., without preconcentration via SPE) and liquid chromatography triple quadrupole mass spectrometry according to a method developed by North Carolina State University (referred to here as Method II). Briefly, 900 μL of the sample was added to a 1.5-mL polypropylene microcentrifuge tube with 100 μL of mass-labeled internal standard mix in methanol. The tube was centrifuged at 15,000 rpm for 15 min at 4°C and 800 μL of the supernatant was transferred to a 2-mL polypropylene autosampler vial. A 200- μL aliquot of each sample was injected onto an Agilent 1290 LC system with a 900- μL sample loop and analyzed on an Agilent 6495c triple quadrupole mass spectrometry system. Due to the

thermally labile nature of some targeted analytes, two separate injections were analyzed to quantitate the compounds with low source temperature or high source temperature settings. Fully specified conditions can be found in Table S4.

Analyte concentrations were calculated with 10-point calibration curves ranging 1–500 ng/L. Calibration standards were analyzed at the beginning and end of the sample batch. Three continuing calibration checks (ranging 10–500 ng/L) were analyzed for every 20 samples. For Method II, the MRL was determined as either (1) the lowest calibration standard detected within $\pm 30\%$ of the true value or (2) the lowest calibration standard where the response exceeded that of the highest method blank by a factor of two. MRLs ranged 1–20 ng/L. A separate MDL was not calculated under Method II.

2.5.3 | Additional water quality analyses

Dissolved organic carbon (DOC) was measured in the influent and effluent samples each month using a Sievers M9 portable total organic carbon (TOC) analyzer. As soon as possible after sample collection, 40 mL was subsampled using a syringe and filtered through a 0.45 μm cellulose acetate membrane filter (Puradisc Aqua, GE Whatman) into baked glass vials. Background levels of DOC from the HDPE bottles after up to 48 h of holding time were determined to be low (< 0.05 mg/L) and did not significantly influence the analysis. Electrical conductivity, pH, and temperature were measured in the field during each sample event using a handheld probe (HI98129, Hanna Instruments, Smithfield, RI) calibrated daily before use.

2.6 | Data analysis

2.6.1 | Statistical tests and tobit regression models

For each household, PFAS removal was assessed by comparing individual and total PFAS concentrations in the filter influent and effluent. PFAS not detected above analytical limits in the filter influent were not included in these analyses. Wilcoxon signed rank tests were used to test for significant differences between influent and effluent concentrations. To estimate percent removal when effluent concentrations were $< \text{MRL}$, mixed-effects Tobit regression models were fit to the log-transformed concentrations of each analyte. This was considered superior to a simple substitution approach where analytes $< \text{MRL}$ or $< \text{MDL}$ are substituted with a constant value since low

levels of PFAS in influent samples close to the MRL or MDL could result in a low estimation of percent removal for certain analytes (Herkert et al., 2020). Thus, a censored regression model was fit to the data according to the equation:

$$C_{i,h}^* = X_{i,h} \beta_{\text{filter}}^* + \mu_h + \varepsilon_{i,h}, \quad (1)$$

where $C_{i,h}^*$ is a latent variable of unobserved (log-transformed) concentrations for observation $i = 1, \dots, N$ clustered in household $h = 1, \dots, N$. $X_{i,h}$ is a vector of independent explanatory variables; β_{filter} is an unknown regression coefficient determining the fixed effect of the filter on the log of the PFAS concentrations; μ_h is a univariate random intercept describing the variance between households; and $\varepsilon_{i,h}$ is an error term (Wang & Griswold, 2016). $C_{i,h}^*$ describes the observed left-censored PFAS concentrations within each household $C_{i,h}$ such that:

$$C_{i,h} = \begin{cases} \ell & \text{if } C_{i,h}^* < \ell \\ C_{i,h}^* & \text{if } C_{i,h}^* \geq \ell \end{cases} \quad (2)$$

where ℓ is the lower limit for each analyte. The lower limit ℓ was set to the MRL as concentrations below this threshold are considered estimated values that do not fully satisfy all quality control objectives of the analytical method (Munch & Branson, 2004).

Using the “lme4cens” and “survival” packages in the software R (Kuhn, 2021; Therneau, 2021), models were fit iteratively for each class of PFAS (all targeted PFAS, PFCAs, PFSAs, and PFEAs) to evaluate the change in PFAS concentration from the filter influent to the effluent. The coefficient β_{filter} was then exponentiated to yield an estimate of the overall percent reduction for each PFAS class such that:

$$\% \text{removal} = 1 - e^{\beta_{\text{filter}}}. \quad (3)$$

2.6.2 | Calculating influent summary statistics

Influent PFAS summary statistics were estimated by maximum likelihood estimation to account for left-censored data below the MDL (Method I) and MRL (Method II) for each analyte. This approach is more accurate and leads to less error than common substitution approaches, especially when the percent of censored data exceeds 15% (Baccarelli et al., 2005; Croghan & Egeghy, 2003). The true mean, median, and a 95% confidence interval around the mean were estimated using the “EnvStats” package in R (Millard, 2013).

Additionally, a common practice during PFAS analysis and risk assessment is to sum the concentrations of all targeted PFAS analytes in each sample. In terms of total PFAS, the sum concentration is uncertain because analytical methods target only a small portion of all possible PFAS, method reporting limits for some PFAS are relatively high, and a concentration of zero is often assigned for analytes below detection or reporting limits. However, summed PFAS concentrations are the basis of new regulatory limits for drinking water quality, such as the 20 ng/L threshold recently established for the sum of five (PFAS₅) and six (PFAS₆) PFAS in Vermont and Massachusetts, respectively (General Assembly of the State of Vermont, 2019; Massachusetts Department of Environmental Protection, 2021). *Consumer Reports* has also begun to independently publicize a recommended threshold of 10 ng/L for the sum of all PFAS (Felton, 2021), and the state of North Carolina implemented a 70 ng/L limit for the sum of 12 PFAS in drinking water surrounding the fluorochemical manufacturer near cluster C in this study (North Carolina General Court of Justice, 2019). Thus, to be able to compare the sum of PFAS in each sample to these emerging thresholds, the sum of detectable PFAS was calculated by substituting MDL/2 (for analytes in Method I) or MRL/2 (for analytes in Method II) for samples below these limits. Summary statistics were calculated from the distribution of sums. This simple substitution approach was compared to a nonparametric Kaplan–Meir estimation for summing nondetect values outlined by Helsel (2010) and was found to closely approximate the true estimate of the sample sum when the left-censoring limit was low.

2.6.3 | Forecasting missing flow data

Due to fieldwork restrictions during the COVID-19 pandemic, sampling visits could not be conducted for 2 months at the end of the study. During this time, the internal memory of the data logger reached capacity, preventing flow data from being collected for each household. To correct for missing data, an additive exponential smoothing forecast model (Figure S3) was used to estimate the cumulative volume of water treated at the end of the study using the “forecast” package in R (Hyndman & Khandakar, 2008).

3 | RESULTS

3.1 | Influent PFAS concentrations

Of the 24 PFAS tested for by Method I, eight analytes (PFBA, PFPeA, PFOA, PFBS, PFPeS, PFHxS, PFOS, and

TABLE 2 Summary of PFAS concentrations above the reporting limit in raw well water representing filter influent

Class	Analyte	Carbon chain length	Analytical method	n	MDL (ng/L)	% >MDL	MRL (ng/L)	% >MRL	Est. true mean (ng/L)	True mean 95% CI (ng/L)	Mean of samples >MRL (ng/L)	Max inf. C (ng/L)
PFCA	PFBA	4	I	82	1.5	63.4	6.2	8.5	4.3	2.9–6.1	33.1	61.7
	PFPeA	5	I	82	0.3	75.6	6.2	1.2	1.9	1.3–2.5	6.8	6.8
	PFHxA	6	I	82	1.6	41.5	6.2	0	1.4	0.9–1.4	NA	4.1
	PFHpA	7	I	82	0.8	39.0	6.2	0	0.8	0.5–0.8	NA	2.9
	PFOA	8	I	82	0.9	54.8	6.2	4.9	2.8	1.7–4.1	7.2	7.4
	PFBS	4	I	82	0.1	98.8	0.5	79	2.9	2.2–3.8	3.4	13
	PFPeS	5	I	82	0.1	89.0	0.5	23	0.4	0.3–0.5	0.9	1.3
	PFHxS	6	I	82	0.2	98.8	0.5	72	1.4	1.1–1.7	1.7	3.9
PFHPS	PFHpS	7	I	82	0.2	11.0	0.5	0	0.1	0.1–0.1	NA	0.4
	PFOS	8	I	82	1.0	29.2	6.2	8.5	12.8	2.7–33.9	15.1	26.3
	HFPO-DA	6	I	82	1.7	70.7	6.2	23	5.3	3.9–6.9	9.0	14.1
	PFMOAA	3	II	9	-	-	5	55.6	5.5	1.6–11	7.8	10.8
	PMPA	4	II	9	-	-	2	100	53.7 ^b	-	53.7	99.5
	PFO2HxA	4	II	9	-	-	1	88.9	4.6	2.1–9.7	4.6	10.1
	NVHOS	4	II	9	-	-	1	77.8	1.1	0.7–1.7	1.3	1.7
	PEPA	5	II	9	-	-	1	100	3.4 ^b	-	3.4	6.0
Nafion BP2	Nafion	7	II	9	-	-	2	88.9	4.3	2.6–7.1	4.6	8.5
	BP2											
\sum PFAS ₁₁				-	-	-	-	-	21.6 ^a	-	-	76.4
\sum PFAS ₁₇				-	-	-	-	-	94.7 ^a	-	-	131.3

Abbreviations: CI, confidence interval; HFPO-DA, hexafluoropropylene oxide dimer acid; MDL, method detection limit; MRL, minimum reporting limit; PFAS, per- and polyfluoroalkyl substances; PFCA, perfluoroalkyl carboxylic acids; PFOA, perfluorooctanoic acid; PFOS, perfluorooctane sulfonic acid; PFSA, perfluoroalkyl sulfonic acids; PMPA, perfluoro-2-methoxypropanoic acid.

^aCalculated value. For analytes in Method I, samples <MDL were substituted with MDL/2; for analytes in Method II, samples <MRL were substituted with MRL/2. PFAS concentrations were then summed for each sample and the mean calculated from the distribution of sums.

^bExact value, no samples <MRL.

HFPO-DA) were detected in the influent above the MRL in at least one study household at least once over the 8-month study duration. Three additional analytes (PFHxA, PFHpA, and PFHpS) were consistently above the MDL in influent samples but never exceeded the MRL (Table 2). These 11 analytes (PFAS₁₁) were included for evaluation of the filter's effectiveness. All wells and all samples had at least one detectable PFAS, and 68 of 82 total influent samples (83%) had at least one PFAS above the MRL. Overall, 61% of individual PFAS₁₁ concentrations in influent samples were above the MDL, and 20% were above the MRL.

Legacy PFOA and PFOS concentrations were below their respective USEPA HALs of 70 ng/L across all three geographic clusters (max = 7.4 and 26.3 ng/L, respectively), but did exceed thresholds put in place by certain states, with 4% exceeding the New Hampshire and Michigan PFOS Maximum Contaminant Levels (MCLs) of 15 and 16 ng/L, respectively (MDEQ, 2020; NH State Legislature, 2020), 7% exceeding the New York PFOS MCL of 10 ng/L (NYDH, 2018), and 5% exceeding the California PFOA Notice Level of 5.1 ng/L (California Water Boards, 2020) and Illinois PFOA HAL of 2 ng/L (IEPA, 2021). PFASs were detected above the MRL more frequently than PFCAs (37% compared to 3% of the time), but the MRL for the PFASs was also lower (0.5 ng/L compared to 6.2 ng/L) due to lower levels of background noise for these analytes. Higher individual analyte concentrations were observed for PFBA (max = 61.7 ng/L), a short-chain PFCA possibly associated with land application of wastewater biosolids (Lindstrom et al., 2011) and landfill leachates (Eschauzier et al., 2013), in cluster B. HFPO-DA, which has been linked to atmospheric emissions from a fluorochemical manufacturer near cluster C (Roostaei et al., 2021), was found at low concentrations in clusters B and C in Robeson County (max = 10.6 and 14.1 ng/L, respectively), but was not detected in any households in cluster A in Orange County over 70 miles north. The sum of influent PFAS₁₁ concentrations among all households ranged 4.7–76.4 ng/L (mean = 21.4 ng/L; Table 2). Overall, PFAS₁₁ concentrations exceeded the Vermont and Massachusetts drinking water threshold of 20 ng/L for the sum of five and six PFAS, respectively, in 34 of 82 samples (41%), and 71% exceeded the *Consumer Reports* recommended threshold of 10 ng/L for the sum of all PFAS.

Nine households in cluster C were also tested for additional PFEAs that have been linked to contamination from the fluorochemical manufacturer (North Carolina General Court of Justice, 2019). These households all had elevated levels of six PFEAs, dominated by perfluoro-2-methoxypropanoic acid (PMPA), which ranged 15.2–99.5 ng/L (mean = 53.7 ng/L; Table 1). The sum of influent

PFAS₁₁ plus the six additional PFEAs (PFAS₁₇) in these households ranged 53.0–131.3 ng/L (mean = 94.7 ng/L). All nine households in cluster C exceeded the legal limit of 10 ng/L for at least one compound established under a North Carolina consent order with the nearby manufacturer, and five of nine exceeded the 70 ng/L limit for the sum of PFEAs (Figure S5) (North Carolina General Court of Justice, 2019).

3.2 | Removal of PFAS₁₁ across filters

Significant removals of PFAS₁₁ were observed across the filters for the entire 8-month follow-up (Figure 2). Due to sampling error by study participants where both samples for a given month were drawn from either the influent or effluent, 18 samples (9 sample pairs or 10% of all samples) were excluded leaving 164 paired samples for analysis, equaling 1084 paired data points when considering all 11 analytes. Of the filter effluent samples ($n = 82$), 4.8% of PFAS₁₁ analytes ($n = 902$) exceeded the MDL and 0.1% were above the MRL. Compared to 61% and 20% of analytes above the MDL and MRL, respectively, in the influent samples, this represents a 92% reduction in the prevalence of detectable PFAS₁₁ and a 99.5% reduction in reportable PFAS₁₁ (i.e., PFAS₁₁ analytes >MRL) over the course of the study. Only one of 82 effluent samples (1.2%) contained any PFAS₁₁ (PFBS) above the MRL, which was detected in the effluent at one household at a concentration <1 ng/L.

Using the assumption that all PFAS <MDL were present at a concentration of MDL/2, the estimated sum of PFAS₁₁ in all effluent samples never exceeded 7.9 ng/L (mean effluent \sum PFAS₁₁ = 4.7 ng/L). Given that substituting MDL/2 for nondetect values is a conservative assumption, the true mean of \sum PFAS₁₁ in effluent samples was likely even lower than this estimate. Paired Wilcoxon signed rank tests on the distributions of influent and effluent samples showed that the effect of the filter on PFAS₁₁ concentrations was highly statistically significant, regardless of chain length (Figure 2). Consistent removal was also observed at each sampling month. The aggregated data in Figure 3 show the overall removal of \sum PFAS₁₁ among all 18 filters over the course of the study, with no significant breakthrough of \sum PFAS₁₁ in the effluent after 8 months (up to 250 days) of use. For comparison, influent and effluent concentrations of individual analytes over time from four households in cluster C are also shown in Figure 4. As can be seen, effluent concentrations were consistently below the MDL throughout the study period. PFBA was detected above the MDL on several occasions in the effluent, suggesting that it may be slightly less well removed than other

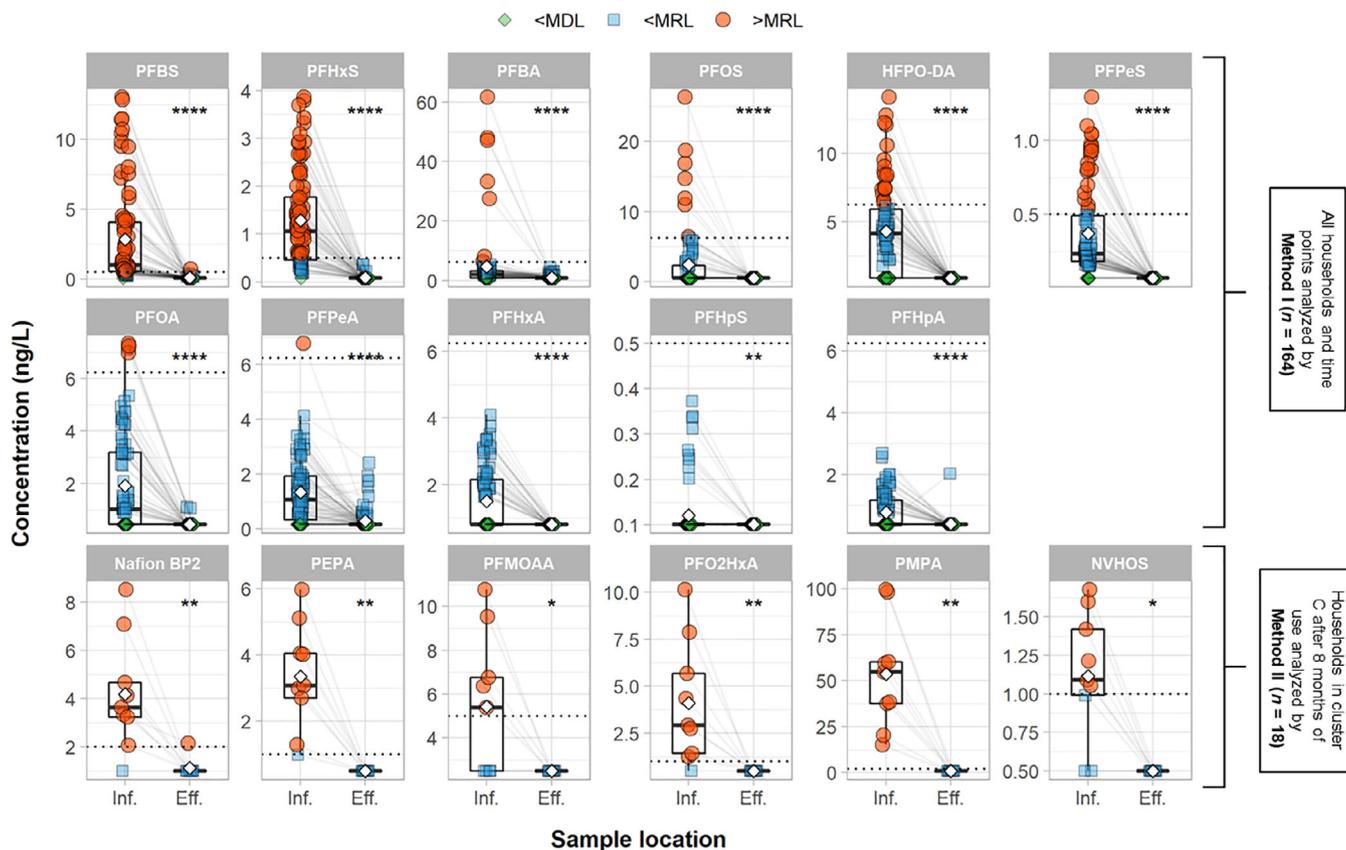


FIGURE 2 Paired filter influent and effluent concentrations of all detectable per- and polyfluoroalkyl substances demonstrating significant reductions in the effluent concentrations for each analyte. Stars indicate significance of difference between influent and effluent samples from Wilcoxon tests. White diamonds show the censored mean. The dotted line in each plot indicates the MRL for each analyte. For plotting purposes, samples <MDL are shown at a concentration of MDL/2. The 11 analytes in the first two rows are aggregated results from all households and time points analyzed by Method I. The bottom row shows results for nine households in cluster C approximately five miles west of a fluorochemical manufacturer after 8 months of use analyzed by Method II. Colors show whether each data point was above the MRL (red), below the MRL (blue), or below the MDL (green). Lines connect paired influent/effluent sample points. MDL, method detection limit; MRL, minimum reporting limit

analytes, but these detections were still below the MRL so were not considered to represent significant breakthrough due to the uncertainty of results below the MRL.

3.3 | Effect of influent water quality and water use on PFAS removal

Variations in influent water quality within the pH range of 4.4–7.1 and DOC range of 0.6–1.2 mg/L (Table 1) did not have a significant effect on ACB treatment effectiveness over the course of the study. Variations in water use by each household during the study also did not influence PFAS removal, even in one household that exceeded the filter's certified volume-based capacity after 8 months of use. The average cold water use at the kitchen tap among all 18 households was 7.6 L/day, representing a cumulative volume of water treated of 210–4655 L (approximately 220–4880 BVs), or only 50% of the filter's

rated capacity on average (mean = 1495 L) (Figures S3 and S4). By comparison, bench- and pilot-scale studies evaluating PFAS removal by GAC observed breakthrough after approximately 5000–50,000 BV of water treated, depending on the carbon type, influent PFAS concentration, and influent DOC (Rodowa et al., 2020; Zeng et al., 2020). Thus, the absence of any effects of influent conditions on treatment effectiveness in this study may primarily be due to the low rates of cold water use at the kitchen tap. This suggests that the lifetime of under-sink ACB filter cartridges (if replaced at the manufacturer-recommended intervals) is likely to fall within the initial lag period of the theoretical S-shaped breakthrough curve exhibited by activated carbon, although testing in additional contexts remains necessary (see considerations for testing in other contexts in Section 4.2 below).

The observed independence from influent pH suggests that hydrophobic rather than electrostatic interactions controlled PFAS removal, as seen elsewhere (Wang

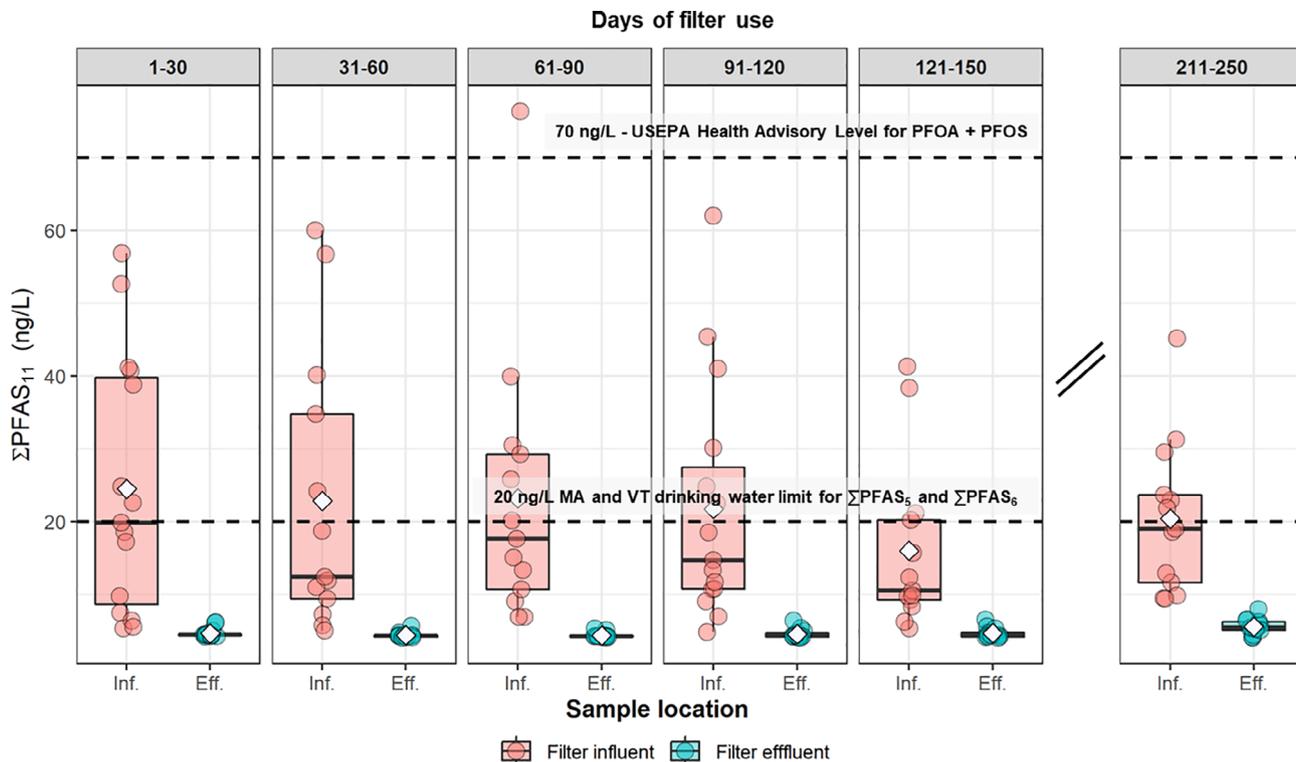
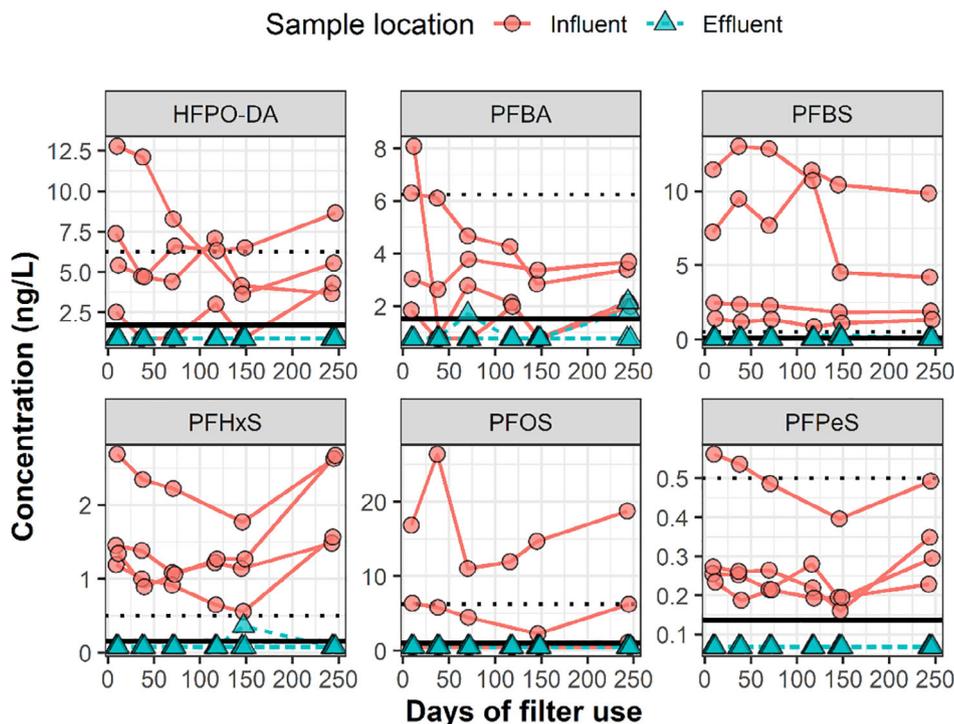


FIGURE 3 Distribution of the sum of PFAS₁₁ concentrations in the influent and effluent of all filters at each sample month compared to existing regulatory thresholds for the sum of five and six different PFAS in Massachusetts and Vermont (20 ng/L) and the USEPA health advisory level for PFOA + PFOS (70 ng/L). Σ PFAS₁₁ was consistently removed for the entire study period with no significant breakthrough observed. PFAS, per- and polyfluoroalkyl substances; PFOA, perfluorooctanoic acid; PFOS, perfluorooctane sulfonic acid

FIGURE 4 Time series influent and effluent concentrations of selected analytes consistently above the MRL for four households in cluster C (analyzed by Method I). Dotted horizontal lines indicate the analyte MRL while solid, horizontal lines indicate the analyte MDL. For plotting, concentrations <MDL are shown at MDL/2. Filters exhibited consistent removal for up to 8 months of use without significant breakthrough. Time series data for additional PFEAs analyzed by Method II were not available but effective removal after 8 months indicates a similar lack of breakthrough for these analytes. MDL, method detection limit; MRL, minimum reporting limit; PFEA, perfluoroalkyl ether acids



et al., 2019; Zeng et al., 2020). Additionally, although the influent DOC levels were low, over 70% DOC breakthrough occurred on average after treatment of <500 BV of water (Figure S6), but no significant PFAS₁₁ broke through during the same period, exhibiting a similar independence from the sorption behavior of bulk organic matter as shown by Zeng et al. (2020) in bench-scale tests.

Although no contaminant breakthrough was detected, clogging of the filter cartridge reduced the flow rate to unusable conditions in 3 of 18 households (17%) after only 2–3 months of use (150–1335 L treated), and the study was ended early for these households. Clogging thus represents an alternative endpoint for the filter's effectiveness and may need to be addressed in households with either very low water use and/or high influent iron or turbidity as discussed elsewhere (Mulhern et al., 2021; Mulhern & MacDonald Gibson, 2020).

3.4 | Removal of emerging PFEAs

The nine filters installed in homes in cluster C removed 98% of influent PFEAs to below reporting limits after 8 months of use (Figure 2). The compound known as Nafion byproduct 2 was detected one time in the effluent of a single household at a concentration of 2.1 ng/L but otherwise was removed to below the MRL. PMPA, which was detected at concentrations 10–50 times higher than other PFEAs on average and three times higher than average PFOA+PFOS concentrations (max = 99.5 ng/L), was removed to below the MRL in all effluent samples. These emerging short-chain PFEAs originating from the fluorochemical manufacturer near cluster C have been shown to be difficult to remove with powdered activated carbon in municipal water treatment scenarios (Sun et al., 2016), but were effectively removed by the under-sink ACB filters without contaminant breakthrough even 2 months beyond the recommended filter lifetime. All effluent samples were well below the North Carolina consent order limit of 10 ng/L for each individual compound and 70 ng/L for \sum PFAS (North Carolina General Court of Justice, 2019).

3.5 | Calculated PFAS removal

The mixed-effects Tobit regression models accounting for left-censored data below the MRL showed that the filter removed 97%–99% of PFAS₁₇ (Table 3). Performance did not vary among the different classes of PFAS, including PFEAs, PFSAs, and PFCAs. The coefficient on the filter term was highly statistically significant ($p < .00001$) for each model except for the model fit to PFCAs due to the

low prevalence of samples with any PFCAs above the MRL in the filter influent (only 1.5%). Regardless, the model estimated 99% removal for this class. In one household where influent samples contained PFBA—a four-carbon PFCa—at levels from 33 to 61 ng/L (mean = 45 ng/L), the filter removed PFBA to below detection in all effluent samples, representing 98% removal on average assuming a value of MDL/2 in the effluent (see facet PFBA in Figure 2).

4 | DISCUSSION

4.1 | Effectiveness of ACB filters for private wells

This study demonstrates that ACB filters can be an effective option to mitigate legacy and emerging PFAS contamination in private well water when used according to the manufacturer's recommendations. Among the 18 households in this study, under-sink ACB filters effectively removed 97%–99% of influent \sum PFAS₁₇ concentrations up to 131 ng/L for the recommended lifetime of the filter cartridge, with no significant differences in removal between individual households. The prevalence of PFAS₁₁ above the reporting limit was reduced by 99.5% and the prevalence of emerging PFEAs above the reporting limit was reduced by 98%. This is the largest longitudinal study of activated carbon-based POU devices for PFAS removal from private well water to date, increasing confidence in the use and testing of these devices to mitigate PFAS in private well water for impacted communities around North Carolina and elsewhere.

The levels of removal shown in this study are comparable to the reported effectiveness of RO filters and two-stage ACB filters in a previous cross-sectional assessment of POU filters to remove PFAS in municipal tap water in North Carolina, where the filters removed >75%–100% of PFSAs, PFCAs, and PFEAs in influent tap waters (influent \sum PFSA+PFCa range: 6–759 ng/L) (Herkert et al., 2020). As was the case here, most compounds were removed to >90%, except for certain analytes for which a minimum removal level was reported because influent levels were close to the method reporting limit. Single-stage under-sink filters in the study by Herkert et al. (2020) were generally not as effective as the filters in this study, which showed an average range of removal from 15% to >99% for 10 PFAAs and from 51% to >99% for seven PFEAs. This wide range of performance is unsurprising given the myriad under-sink filter technologies on the market, from simple sediment and chlorine removal filters to ion exchange resins and activated carbon, and the relative paucity of products that are certified

TABLE 3 Calculated percent removal across the filter for each PFAS type from mixed effects Tobit regression models

Analyte	<i>n</i>	% left censored	β	β 95% CI	<i>p</i> -value	Est. % removal	% removal 95% CI	% >MRL in effluent
All PFAS ₁₇	1912	88.1%	-4.22	-5.00 to -3.43	<.0001	99%	97%-99%	0.2%
PFEAs	272	76.5%	-3.50	-4.52 to -2.48	<.0001	97%	92%-99%	0.2%
PFSAs	820	81.6%	-3.91	-4.80 to -3.02	<.0001	98%	95%-99%	0.7%
PFCAs	820	98.5%	-6.65	-	.95	99%	-	0%

Note: *N* indicates the total number of paired influent and effluent samples included in the regression model. Tobit regression model was not significant for PFCAs due to low prevalence of PFCAs in the filter influent above the MRL.

Abbreviations: MRL, minimum reporting limit; PFAS, per- and polyfluoroalkyl substances; PFCA, perfluoroalkyl carboxylic acids; PFEA, perfluoroalkyl ether acids; PFSA, perfluoroalkyl sulfonic acids.

for PFAS removal under NSF P473. The same is true of refrigerator and pitcher filters, which were also shown to be less effective for PFAAs (only 29%–72% and 36%–71% removal, respectively) and PFEAs (56%–>99% and 46% removal, respectively) (Herkert et al., 2020). Faucet-mounted filters showed slightly better removal of PFAAs (63%–99%) but still not as high as in this study. Thus, more advanced filters with a two-stage filtration process utilizing activated carbon and/or single-stage filters with a large-volume ACB (such as the device tested in this study) are more likely to be effective for PFAS removal from both municipal tap water and private well water. NSF P473 certification may be a better indicator of effectiveness for PFAS than device design (i.e., under-sink, two-stage, single-stage), although certification for PFOA +PFOS removal does not necessarily guarantee removal of short-chain PFAAs and PFEAs in other contexts, despite their being well-removed in this study.

Differences in removal reported by Herkert et al. (2020) compared to this study may also be due to prolonged use of the filters beyond the manufacturer recommended replacement interval. If users do not replace the filter cartridge at the recommended intervals, they may be at risk of PFAS breakthrough, with short-chain PFAS and less strongly adsorbing PFCAs (e.g., PFBA) more likely to break through first (Rodowa et al., 2020; Zeng et al., 2020). Indeed, in laboratory tests conducted by the Minnesota Department of Health with faucet-mounted ACB filters, PFBA was the first analyte to be detected above the detection limit in the filter effluent after 75% of the filter's capacity had been reached (MDH, 2017). Although no breakthrough was observed among the filters in this study, most households had not reached the manufacturer's volume-based capacity recommendation even after 8 months. The exact time-to-breakthrough thus critically depends on each household's rate of water use. The results of this study suggest that the manufacturer-recommended lifetime provides a significant safety factor in its estimates of cold-water tap use

over 6 months and should be followed carefully in order to minimize the risk of breakthrough exposures.

Although RO systems are generally considered the safest option for both municipally treated and private well water, ACB filters may appeal to many private well users given that they are simpler to maintain than RO systems, generally have lower capital, operation, and maintenance costs (USEPA, 2007), generate significantly less waste, and use less water (USEPA, 2006). RO membranes have also been shown to degrade rapidly in household contexts (Pratson et al., 2009), and POU RO devices have exhibited sporadic breakthrough of up to 25% of influent PFAS during laboratory testing, potentially due to leaking membrane seals or poor prefilter performance (Patterson et al., 2019). Three of 18 ACB filters in this study clogged prematurely; however, suggesting that POU or point-of-entry sediment prefilters may be necessary to extend the capacity of ACB filters for some private well waters. ACB filters may thus be a valid alternative to RO for PFAS depending on influent water quality and PFAS concentrations. State and local public health authorities charged with providing technical guidance to well owners ought to consider the tradeoffs of each filter type according to the local context.

4.2 | Considerations for application in other contexts

The effectiveness of ACB filters may still vary under different conditions. Higher rates of water usage, influent PFAS concentrations, and influent DOC may exhaust the carbon capacity more rapidly and increase the possibility of early breakthrough. For example, Anumol et al. (2015) showed that two different ACB refrigerator filters treating groundwater with low influent organic matter were able to remove >97% of PFOA and PFOS for the entire manufacturer estimated lifetime, but the same two devices treating surface water with higher TOC (up to 3.2 mg/L)

showed significantly reduced performance, with 60% breakthrough of PFOA occurring in one filter after treating only half of the filter's rated capacity. Thus, a range of performance may be expected for the same product treating different influent water types. The wells in this study had low organic matter (<1 mg/L DOC on average; Table 1) compared to surficial aquifers in coastal North Carolina with an average influent DOC of 2.5 mg/L (median = 1.5 mg/L, maximum = 9.2 mg/L) (Harden et al., 2003). Other groundwaters may have DOC levels of less than 1 mg/L to over 20 mg/L (Gooddy & Hinsby, 2009). Shallow aquifers and aquifers close to surface water generally have higher DOC levels and thus may be more challenging to treat using ACB filters (McMahon et al., 2019). NSF P473 certification requires devices to be challenged with >1 mg/L TOC in the influent, but pilot testing in additional communities remains essential to inform context-specific performance.

Household testing with NSF P473-certified products under higher influent concentrations should also be piloted. Indeed, the groundwater concentrations in this study (max $\sum \text{PFAS}_{17} = 131.2$ ng/L) were low compared to other communities near PFAS sources where PFOA concentrations alone have been reported at levels of 1800 ng/L up to 20,000 ng/L (Hoffman et al., 2010; Patterson et al., 2019; Xiao et al., 2015). ACB filters—and POU water treatment in general—may not be appropriate for these high-end scenarios. However, NSF P473 requires filters to be challenged with 1500 ng/L PFOA + PFOS (NSF Joint Committee on Drinking Water Treatment Units, 2016), over 11 times the highest $\sum \text{PFAS}_{17}$ concentration tested in this study. Although NSF P473 does not consider PFEAs or short-chain PFAAs, this comparison suggests that the filter could also be effective for well water with significantly higher influent PFAS than tested in this study.

4.3 | Potential for PFAS exposure reduction

Many private well users may be chronically exposed to low PFAS concentrations even in areas without acute point sources or histories of industrial PFAS use due to diffuse applications of AFFF, leaching from septic systems, and/or rural applications of biosolids (Lee & Murphy, 2020). One effort to map PFAS exposure risks from groundwater in Rhode Island suggests that the highest risk regions are likely to be rural areas that have a lower density of PFAS sources but are more vulnerable to groundwater contamination and have likely exposure routes through private and small community wells (Guelfo et al., 2018). Two studies of private drinking water

wells distant from industrial point sources in Cape Cod, Massachusetts, for example, detected individual PFAS concentrations up to 97 ng/L, with approximately half of the wells above detection, from the impact of onsite wastewater systems alone (Schneider et al., 2014, 2016).

Although some nationwide data are available for PFAS in public water supplies in the United States following the USEPA's Unregulated Contaminant Monitoring Rule 3 (UCMR3), no nationally representative data set currently exists for estimating ambient PFAS levels in private well water (Hu et al., 2016; Lee & Murphy, 2020). Of all samples containing detectable PFAS in the UCMR3 results, 72% originated from systems served by groundwater which had an average concentration of 210 ng/L for the sum of six PFAS (Guelfo & Adamson, 2018). A national screening of 163 raw groundwater sources in France also found levels from 1 to 62 ng/L for 10 different PFAS, comparable to the levels in this study (Boiteux et al., 2012).

In these scenarios, under-sink ACB filters could be effectively deployed to reduce chronic PFAS exposures and adverse health outcomes among private well users. Predictive modeling and machine-learning may also be used to identify at-risk areas that may benefit from POU treatment interventions (Hu et al., 2021; Roostaei et al., 2021). Research has shown that extended exposure to low levels of PFOA and PFOS in drinking water can lead to a 100-fold or greater increase in blood serum levels (Emmett et al., 2006; Hoffman et al., 2010; Hurley et al., 2016; Post et al., 2012). At this drinking water: serum ratio, even the low influent PFOA and PFOS concentrations in this study (Table 2) could result in blood serum levels up to 20 times higher than the serum-based reference dose of 0.1 ng/mL proposed by Grandjean and Budtz-Jørgensen (2013) based on reduced vaccine response in children. As research regarding an appropriate reference dose continues for many PFAS (Brown et al., 2020; Guelfo et al., 2018), these data suggest that implementation of ACB filters could help reduce immunotoxic and other health effects associated with chronic, low-level exposures to PFAS from private well water.

5 | CONCLUSION

5.1 | Taking proactive measures to protect well users

Communities addressing PFAS contamination worldwide are challenged by a lack of information on sources, health effects, and fate and transport of legacy and emerging PFAS in the environment (Guelfo et al., 2018).

These knowledge gaps prevent consensus building and effective risk management at federal, state, and local levels. Municipal drinking water utilities may assume significant costs for treatment upgrades and long-term strategic planning to anticipate the evolving scientific and regulatory landscape (Whitby et al., 2021), but private well users generally lack the financial support and extensive technical expertise required to comprehensively evaluate potential PFAS sources, assess risk, conduct advanced testing, interpret toxicological data, and design a proactive treatment or mitigation plan. Thus, when it comes to PFAS, the fundamental question for many well users—“Is my water safe to drink?”—is fraught with uncertainty (Harclerode et al., 2021; Wickham & Shriver, 2021). Rather than waiting for legislative bodies to “catch up” with the scope of PFAS contamination and emerging toxicological data to provide this clarity and recommend action, local governments and community organizers should take practical measures to protect private well users even as research and regulatory decisions are ongoing. As one such practical step, numerous researchers have called for improved information regarding the effectiveness of available treatment interventions to guide public health authorities and community stakeholders in making evidence-based recommendations for well users (Guelfo et al., 2018; Harclerode et al., 2021; Lee & Murphy, 2020; Seltnerich, 2019). The results of this study suggest that under-sink ACB filters certified under NSF P473 could provide a relatively low-cost intervention for private well users. Pilot studies of similar ACB devices may thus be conducted with greater confidence in other communities to determine the effectiveness within each context (e.g., higher influent concentrations, different PFAS species, higher dissolved organic matter, different pH range). Individuals may also use these results to make informed decisions about water treatment in their own home, although robust technical and financial support systems are also necessary to effectively implement POU water treatment programs among well users (Mulhern et al., 2022). State and local environmental and public health authorities may thus use this information to provide proactive support to communities on private wells that may be at risk of chronic PFAS exposures.

ACKNOWLEDGMENTS

The authors thank Bill Smith, Melissa Packer, Michele Fanning, and Cedric Locklear at the Robeson County Health Department and Rev. Robert Campbell at the Rogers Eubanks Neighborhood Association for support during participant recruitment. Field work and sampling support were provided by Hania Zanib, Tejas Pruthi, Banks Grubbs, Jared Peck, April Desclos, Megan Stallard, and Emanuele Sozzi at UNC Chapel Hill. Brad Gibson

and Glenn Walters supported the treatment system design and installation. The authors thank Josh Kearns for providing the carbon analysis. This work was supported by the North Carolina Policy Collaboratory at the University of North Carolina at Chapel Hill.

CONFLICT OF INTEREST

The authors report no conflicts of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

AUTHOR CONTRIBUTIONS

Riley Mulhern: Conceptualization; data curation; formal analysis; investigation; visualization; methodology; writing – original draft; project administration. **Nichole Bynum:** Investigation; methodology; writing – review and editing. **Chamindu Liyanapatirana:** Resources; methodology; writing – review and editing. **Noelle J. DeStefano:** Investigation; methodology; writing – review and editing. **Detlef R. U. Knappe:** Resources; supervision; methodology; writing – review and editing. **Jacqueline MacDonald Gibson:** Conceptualization; resources; supervision; funding acquisition; project administration; writing – review and editing.

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How to cite this article: Mulhern, R., Bynum, N., Liyanapatirana, C., DeStefano, N. J., Knappe, D. R. U., & MacDonald Gibson, J. (2021). Longitudinal assessment of point-of-use carbon filters for removal of per- and polyfluoroalkyl substances from private well water. *AWWA Water Science*, e1262. <https://doi.org/10.1002/aws2.1262>