POINT-OF-USE WATER TREATMENT FOR PRIVATE WELLS IN NORTH CAROLINA: RISKS AND SOLUTIONS FOR LEAD, PER- AND POLYFLUOROALKYL SUBSTANCES (PFASs), AND MICROBIAL CONTAMINANTS

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ABSTRACT

Riley E. Mulhern: Point-of-Use Water Treatment for Private Wells in North Carolina: Risks and Solutions for Lead, Per- and Polyfluoroalkyl Substances (PFASs), and Microbial Contaminants
(Under the direction of Jacqueline MacDonald Gibson)

Almost 50 million people across the United States and Canada rely on privately-owned wells for their domestic water needs. Private well users are legally responsible for maintaining their own water quality as no enforceable drinking water standards nor monitoring and treatment requirements exist for private wells as they do for public water systems. As a result, private well users are potentially exposed to a range of chemical and microbial contaminants through their drinking water. Point-of-use (POU) water treatment represents one potential solution to reduce harmful exposures through well water, but well users frequently do not adopt any household treatment even after the results of a water test indicate some form of contamination. Additionally, the effectiveness of consumer POU treatment products for removing certain contaminants from private well water, including lead and per- and polyfluoroalkyl substances (PFASs), is largely unknown, as very few studies have tested their performance over time in household settings. This research thus aims to advance knowledge around POU water treatment for private well water to reduce the barriers to these solutions for well users. An under-sink activated carbon block POU filter was installed in the homes of 18 well users across North Carolina and tested monthly for eight months for metals, PFASs, and microbial indicator organisms. Filters removed 98% of all influent lead on average for the entire study duration and significantly improved the safety and effectiveness of faucet flushing, reducing levels at the tap to below the American Academy of Pediatrics recommendation of 1 µg/L of lead in drinking

water. Additionally, filters consistently removed 97-99% of all influent PFASs, including emerging short-chain perfluoroalkyl ether acids, even up to two months beyond the manufacturer recommended lifetime of the device. Filters did not result in increased microbial risk of drinking water at the tap under normal conditions of use, but the results emphasized the need for additional well protections and maintenance to ensure the microbial safety of private well water, regardless of the decision to implement POU treatment. Finally, the experiences of participants in this study highlighted the need for strengthened well outreach and support programs that provide technical assistance and financial support to private well users around POU treatment in addition to well testing. The results of this study may thus be used by state and local health departments to provide evidence-based recommendations around the use of POU filters for private well users that may significantly reduce lead and PFAS exposures among this population.

For everyone who gets their water from a well.

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PREFACE

In the words of the theologian Karl Barth, “No act of man can claim to be more than an attempt, not even science…[We] can do no more than question after the better, and never forget that we are succeeded by other, later men; and he who is faithful in this task will hope that those other later men may think and say better and more profoundly what we were endeavoring to think and to say.” I hope that these pages may both serve to some practical degree the men and women of today, as well as inspire other men and women after me to improve upon this attempt.

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LIST OF ABBREVIATONS

AC	Activated carbon
AC-POU	Activated carbon point-of-use
ACB	Activated carbon block
AFFF	Aqueous film forming foam
AGI	Acute gastrointestinal illness
ANSI	American National Standards Institute
ATSDR	Agency for Toxic Substances and Disease Registry
BV	Bed volume
CDC	Centers for Disease Control and Prevention
CFU	Colony forming units
CSMR	Chloride sulfate mass ratio
DOC	Dissolved organic carbon
GAC	Granular activated carbon
HDPE	High density polyethylene
HPC	Heterotrophic plate count
HWISE	Household Water Insecurity Experiences Scale
ICP/MS	Inductively coupled plasma mass spectrometry
IRB	Institutional Review Board
LC	Liquid chromatography
LCMRL	Lowest concentration minimum reporting limit
LOQ	Limit of quantitation
LSI	Langelier saturation index

MDL	Method detection limit
MPN	Most probable number
MRL	Minimum reporting limit
NCDEQ	North Carolina Department of Environmental Quality
NGWA	National Ground Water Association
NSF	National Sanitation Foundation
NTU	Nephelometric turbidity units
PFAS	Per- and polyfluoroalkyl substance[1]
PFU	Plaque forming units
Pb	Lead
POU	Point-of-use
RO	Reverse Osmosis
SDWA	Safe Drinking Water Act
SPE	Solid phase extraction
TOC	Total organic carbon
TSA	Tryptic soy agar
TSB	Tryptic soy broth
UCMR	Unregulated Contaminant Monitoring Rule
USEPA	United States Environmental Protection Agency
USGS	United States Geological Society
VOC	Volatile organic compound
WHO	World Health Organization

[1] For a complete list of the chemical names of individual PFASs, the reader is referred to Appendix B – Table B.3.

CHAPTER 1: INTRODUCTION

1.1 Summary of Research Objectives

The goal of this research is to inform public health policy and practice regarding the effectiveness of point-of-use (POU) water treatment solutions for private, unregulated water supplies in the U.S. and Canada. Specifically, this work responds to four gaps in knowledge surrounding POU water treatment that prevent the implementation of evidence-based and policy-driven solutions at scale, including:

- Lack of data to demonstrate the effectiveness of POU water filters to mitigate elevated lead levels in drinking water from private wells;
- Incomplete knowledge of the longitudinal performance of POU water filters to remove the emerging organic contaminants known as per- and polyfluoroalkyl substances (PFASs) from private well water;
- Understudied impacts of microbiological changes in water quality as a result of POU water treatment and possible unintended risks;
- Inadequate understanding of user experience and perceptions around POU water treatment that drive household stewardship behaviors.

These knowledge gaps cause considerable confusion regarding the utility of POU water treatment for private wells specifically, and decentralized water treatment approaches broadly, restricting decision-making to adequately respond to water quality risks in private well water at governmental and household levels. In response, this research is structured around the following objectives:

1. Verify the long-term performance of POU filters for lead removal as a strategy to mitigate childhood lead poisoning among households reliant on private wells.
2. Conduct the first longitudinal evaluation of activated-carbon based POU water filters for the removal of PFASs from groundwater and relate observed PFAS removal and breakthrough to chemical structure, carbon characteristics, water quality, and household water usage patterns.
3. Monitor the microbial water quality of POU filter effluent in parallel with chemical removal over time in order to evaluate the microbial health protectiveness and/or risks associated with POU filter use for untreated, undisinfected well water.
4. Assess the beliefs and perceptions of well users regarding POU water treatment before and after the intervention to inform effective outreach to private well users.

1.2 Background and Motivation

1.2.1 The Challenge of Private Water Supplies

Over 42 million people in the United States (U.S.) and 4 million people across Canada rely on privately-owned wells for their domestic water needs (Dieter et al., 2018; Statistics Canada, 2017). Under the Safe Drinking Water Act (SDWA) in the U.S., no legally enforceable drinking water standards nor monitoring and treatment requirements exist for private wells as they do for public water systems, i.e., community water systems with more than 15 service connections. In Canada, the federal government provides national guidelines for drinking water quality, but private wells are also unregulated at the provincial level (Lee and Murphy, 2020). Consequently, private well users are legally responsible for maintaining their own water quality, requiring significant financial resources and technical knowledge, often necessitating professional help. In essence, all the same responsibilities that municipal water systems assume to ensure safe drinking water quality, including source water protection, regular monitoring, treatment, and maintenance, fall on the shoulders of individuals.

Due to a lack of knowledge, information, and resources among well users and fragmented or non-existent private well support programs (Charrois, 2010; Gibson and Pieper, 2017), this burden represents a significant public health risk for this population. Although the perception that groundwater is naturally free from contamination and/or cleaner than municipal water supplies is common among well users (Fizer et al., 2018; Thomas et al., 2019), it is largely unfounded. Poorly constructed or maintained wells can allow seepage of contaminated surface water directly into the aquifer during precipitation events (Simpson, 2004). Even in properly constructed wells, myriad mechanisms of groundwater contamination exist that can introduce microbiological and chemical risks, including agricultural inputs, septic system leachate, landfills and industrial sites, as well as natural and geogenic sources (Balazs et al., 2011; Charrois, 2010; Hepburn et al., 2019; Hoffman et al., 2010; Lee and Murphy, 2020; Lindstrom et al., 2011; Schaider et al., 2016). Certain contaminants, such as lead, copper, and other metals, may originate from household plumbing components that are more easily mobilized in private well water than municipally treated water supplies (Belitz et al., 2016; Pieper et al., 2015, 2018). As a result, well water quality across the U.S. and Canada can be highly uncertain, especially in industrial and agricultural regions. In a national assessment of groundwater quality, over one in five drinking water wells in the U.S. had a chemical contaminant of health concern above the recommended limits, and approximately one third of wells contained some form of microbial contamination (DeSimone et al., 2009), potentially exposing millions of people across the U.S. and Canada to harmful contaminants in drinking water every day.

Research has also established links between private well water and adverse health outcomes. Regarding microbial risks, approximately 59% of all waterborne disease outbreaks in small, non-community drinking water systems across the U.S. and Canada between 1971 and 2014 were associated with consumption of contaminated, untreated, or inadequately treated groundwater (Pons et al., 2015). Similarly, 31% of all waterborne disease outbreaks identified in the U.S. between 1971 and 2006 were associated with consumption of untreated groundwater (Craun et al., 2010). Murphy et al. (2016) estimate that 78,000 cases of acute gastrointestinal illness (AGI) may be attributable to private well use in Canada. Others have estimated that 22% of annual emergency department visits for AGI and $40 million in medical costs could be attributable to microbial contamination of private wells in North Carolina (Defelice et al., 2016; Stillo and MacDonald Gibson, 2017).

Well users can also be disproportionately exposed to chemical risks through their drinking water. A recent study of 59,483 blood lead records from children in in Wake County, North Carolina from 1985 to 2017 demonstrated that children who rely on private wells for domestic needs exhibit a 25% increased odds (95% CI 6.2–48%, $p<0.01$) of elevated blood lead levels than children connected to regulated public water systems (Macdonald Gibson et al., 2020). Similarly, a statewide assessment of over 688,000 infants born between 2003 and 2008 found a significant association between elevated manganese concentrations in private well water and increased prevalence of conotruncal heart defects at birth (prevalence ratio=1.6, 95% CI: 1.1–2.5) (Sanders et al., 2014). Although less common, cases of infant methemoglobinemia (i.e., "blue baby syndrome") have been connected to elevated nitrate levels from private well water used to prepare infant formula (Knobeloch et al., 2000). Private well users in Ohio and West Virginia living near an industrial fluorochemical manufacturer have also been shown to exhibit elevated levels of perfluorooctanoic acid, a synthetic chemical associated with potential reproductive effects, cancer risk, and liver damage, in their blood serum 20 times higher than the average U.S. population (Hoffman et al., 2010).

What is more, racial minorities and low-income communities in the U.S. that are reliant on private wells may experience disproportionate drinking water exposures from contaminants such as lead (Stillo and MacDonald Gibson, 2017, 2018), pathogens (Bischoff et al., 2012; Mattos et al., 2020; Rowles et al., 2020; Stillo and MacDonald Gibson, 2017), arsenic (Rowles et al., 2020), and nitrate (Balazs et al., 2011; Schaider et al., 2019) as a result of historical and ongoing processes of exclusion from municipal services and infrastructure (VanDerslice, 2011). Examples of structural exclusion from municipal services and water access have been documented throughout the U.S, including municipal underbounding in areas of the South (Aiken, 1987; Flowers et al., 2019; Heaney et al., 2015; Lichter et al., 2007; Lockhart et al., 2020; MacDonald Gibson et al., 2014; Naman and Gibson, 2015), redlining and selective annexation practices in California (Balazs and Ray, 2014; Seaton and Garibay, 2009), housing discrimination in the Midwest (Colfax, 2009), and informal *colonias* along the U.S.-Mexico border (Durst, 2019; Rowles et al., 2020). More broadly, people of color have been shown to be 35% more likely to lack water service across the U.S. as compared to white, non-Hispanic households (Meehan et al., 2020).

Ensuring the safety of private water supplies is thus a pressing public health, engineering, and environmental justice issue. Although it may be feasible to extend water lines to unserved areas near municipal limits (see Heaney et al., 2015; Lockhart et al., 2020), connecting to municipal distribution systems is unrealistic for well users in rural areas. Bottled water is also widely available to private well users as a temporary solution, but it does not address the underlying problem of well water contamination and, as discussed briefly in Chapter 5, may place an inequitable economic burden on well users. Thus, current research revolves around minimizing the inherent risks and vulnerabilities associated with private well use (Flanagan et al., 2018; Kreutzwiser et al., 2011; Malecki et al., 2017). In the absence of direct legislation that dedicates public resources to meeting these challenges, a systems-level approach is required to broadly promote and facilitate health protective well stewardship behaviors, which follow a cyclical pattern of testing, interpretation, treatment, and maintenance (**Figure 1.1**). Multiple stakeholders must be involved in creating a sustainable "infrastructure for stewardship" including enhanced individual, county, and state-level capacity for well testing, monitoring, and data collection, promotion of well inspection and maintenance guidelines (Kreutzwiser et al., 2011; Simpson, 2004), and increased use of decentralized household and POU water treatment interventions (Fox et al., 2016).

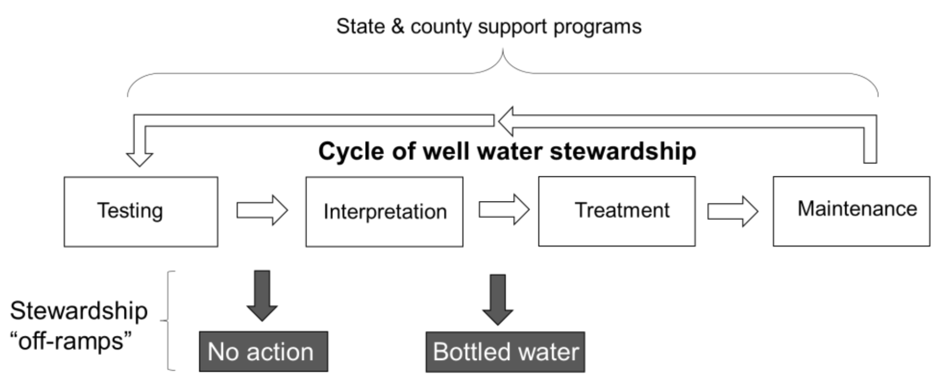

Figure 1.1. Cycle of well water stewardship and frequent "off-ramps" that well users take at critical junctures.

Significant social, economic, regulatory, and technological challenges remain for each of these foci of well stewardship, however, each deserving of in-depth research inquiries. A large body of research has been dedicated to promoting private well water testing as the initial starting point to greater stewardship, i.e., the “on-ramp” or external cue (Colley et al., 2019), but multiple “off-ramps” exist that prevent well users from continuing long-term health protective behaviors, including a failure or inability to interpret the results of water tests, and failure of water test results to translate to well stewardship behaviors, such as conducting a visual wellhead inspection or adoption of water treatment, even when contaminants are present (**Figure 1.1**). For example, in a survey of 386 well users in Maine who had previously been notified that their well exceeded the safe limit for arsenic in drinking water, 27% of well owners reported taking no action after receiving the results, and 30% simply chose to switch to bottled water rather than adopting treatment (Flanagan et al., 2015). Similarly, only 26% of well users in a survey of 1,567 well users in Ontario, Canada reported installing a treatment system in response to contamination (Kreutzwiser et al., 2011).

Thus, when contaminants are present, the crux of the cycle in **Figure 1.1** is the step between testing/interpreting the water test results and taking appropriate mitigative actions in the form of treatment or well maintenance. Several important questions emerge, including, what prevents adoption of treatment? Can household water treatment be safe and effective for private well water? And, how can the infrastructure for stewardship be strengthened to minimize these off-ramps? This research aims to address each of these questions to advance knowledge and improve practice around treatment, specifically addressing the effectiveness and feasibility of POU water treatment, to inform the design of holistic interventions for private well users in the U.S. and Canada.

1.2.2 The Challenges of Point-of-Use Treatment

Decentralized POU water treatment, which by definition is a practice or technology that can be employed to make water safe to drink at the point where it is consumed rather than at a centralized water treatment facility, has been implemented worldwide as a practical means to increase access to safe drinking water among low-income countries, with an emphasis on reducing diarrheal disease (Sobsey et al., 2008). With increasing concerns surrounding the safety of drinking water in high-income countries, however, POU water treatment has also become commonplace as a control strategy for chemical risks associated with the distribution of municipal water supplies such as lead, disinfection byproducts, and trace organic contaminants (Anumol et al., 2015; Brown et al., 2017; Cotruvo and Cotruvo, 2003). Especially in small systems, where the costs of advanced treatment to address emerging drinking water contaminants may be prohibitively high for the customer base to assume, POU treatment has emerged as an affordable means to achieve compliance with national drinking water standards (USEPA, 2006).

The public adoption of POU devices has been accelerated in the U.S. by widely publicized events such as the Washington D.C. and Flint, Michigan lead crises in 2001 and 2015, and concerns around PFASs following the U.S. Environmental Protection Agency's (USEPA) announcement of health advisory levels for the chemicals perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) in drinking water in 2016 (USEPA, 2016a, 2016b). Indeed, nearly 40% of U.S. consumers are estimated to use some sort of household water treatment device (Anumol et al., 2015). In 2018, the POU water treatment market was valued at $19.9 billion and is expected to reach over $30 billion by 2026, largely driven by increasing awareness around chemical quality of drinking water supplies (Reports and Data, 2019).

Despite the size of the industry and widespread use of these technologies across the country, significant knowledge gaps remain regarding their utility and effectiveness for private well owners, including: 1) a lack of evidence demonstrating their effectiveness for high priority chemical contaminants in well water such as lead and PFASs; 2) uncertainty regarding the microbiological safety of common POU filter designs for undisinfected private wells; and 3) incomplete understanding of the factors and perceptions among private well users that drive decision-making and behavior change.

First, the vast majority of research addressing POU water treatment among private well users in the U.S has focused on arsenic removal (Borja-Cacho and Matthews, 2008; Flanagan et al., 2015; George et al., 2006; Möller et al., 2009; Powers et al., 2019; Slotnick et al., 2006; Thomas et al., 2019; Walker et al., 2008; Zheng, 2017), but additional chemical contaminants need to be considered in designing appropriate POU interventions for private well users in certain circumstances. Specifically, despite widespread distribution of water filters for lead removal in the wake of municipal lead crises, no previous U.S. studies have characterized the effectiveness of POU filters for lead removal from private well water over time, leaving well users without evidence-based information to make decisions to reduce waterborne lead exposure. The effectiveness of POU treatment for lead removal from private well water is addressed in Chapter 2.

The class chemicals known as PFASs is another possible contaminant in private well water for which there is a dearth of research on the effectiveness of POU treatment. PFASs have been detected in groundwater throughout the country due to their widespread use in myriad industrial and consumer applications for the past eight decades (Guelfo and Adamson, 2018; Hu et al., 2016). Of the thousands of PFASs in production and potentially hundreds present in environmental media, the National Sanitation Foundation (NSF), an independent organization that sets standards and provides certifications of contaminant reduction claims for consumer water treatment devices, has developed a POU device certification encompassing only two legacy compounds (PFOA and PFOS under NSF P473), leaving open questions for well users around the protectiveness of POU devices against emerging PFASs detected in U.S. water supplies (Sun et al., 2016). Even for NSF-certified products, the variability in private well water quality may represent a range of treatment effectiveness. The performance of POU filters for the removal of PFASs in relation to water quality characteristics and household usage patterns is explored in Chapter 3.

Second, most POU treatment technologies currently available to consumers in the U.S. and Canada are tailored to the removal of chemical contaminants alone and are designed for consumers on municipal water supplies who have microbiologically safe drinking water, rather than for private well users. The 1996 Amendments to the SDWA (Section 1412(b)(4)(E)(ii)), which allow small public water systems in the U.S. to use POU devices for compliance with certain chemical maximum contaminant levels, explicitly prohibit the use of POU technologies to achieve compliance for microbial water quality. The USEPA and World Health Organization have also recommended that activated carbon POU filters not be used with private water supplies where the influent water is not guaranteed to be microbiologically safe (USEPA, 2006; WHO, 2003). Recent research shows that POU filters using activated carbon, a common filter media for water treatment, can be colonized by potential pathogens even in disinfected municipal water supplies (Chaidez and Gerba, 2004; Su et al., 2009; Wu et al., 2017). Thus, significant uncertainty exists around the microbiological safety of commonly used activated carbon POU devices for private well users, deterring their use even where they may be highly effective for chemical contaminants. Chapter 4 discusses the microbial safety and potential risks of activated carbon POU treatment of private well water.

Lastly, a growing body of research exists surrounding the motivations and beliefs driving increased water testing behavior among well owners (Colley et al., 2019; Morris et al., 2016; Paul et al., 2015; Renaud et al., 2011; Stillo et al., 2019; Straub and Leahy, 2014), but the field has only recently begun to address the cognitive factors that motivate or hinder adoption of water treatment (Flanagan et al., 2015; Malecki et al., 2017). As a behavioral intervention, lack of technical knowledge and self-efficacy among well users may limit the utility of these devices to prevent harmful exposures and improve water security at the household level. As one of the earliest scholarly papers on modern POU treatment technologies, published in 1914, astutely pointed out before its time:

> *Any manufacturer of household filters who has not yet learned this lesson must come to recognize that it is impossible to produce an entirely fool-proof filter and that the most ingenious and scientifically correct types will shipwreck on the human element which enters into the supervision and care required to retain continuous efficiency. Directions are worthless, or practically so, unless they are followed in more than a perfunctory manner* (Lederer and Bachmann, 1914).

Over a century later, understanding this "human element" continues to be essential to evaluating the overall effectiveness of POU technologies for private well users. This study was not designed to elucidate behavioral or cognitive questions experimentally, but observational data of the perceptions and experiences of well owners using POU water treatment is an important starting point to shape effective programs and outreach for well owners in the future. Considerations of user experience, perceptions, and beliefs are addressed in Chapter 5.

Each of these areas of inquiry—the technical engineering assessment of treatment effectiveness (Chapters 2 and 3), evaluation of microbial risk (Chapter 4), and analysis of well users' perceptions of POU treatment (Chapter 5)—provide practical, actionable results that may be used to inform the development of effective, evidence-based strategies for safer drinking water among private well owners. Potential implications and future outcomes of this research are discussed in the context of each chapter and summarized in Chapter 6.

1.3 Overview of Research Approach

1.3.1 Study Design

The following chapters are based on a longitudinal assessment of the real-world effectiveness of consumer POU water filters for a sample of 18 private well users in North Carolina. North Carolina has one of the largest concentrations of domestic well users in the U.S. by absolute number (2.4 million) and proportion (24%) (Dieter et al., 2018). Well users were recruited in collaboration with community partners in Orange County and Robeson County in close proximity to potential environmental hazards, including industrial animal farms, municipal landfills, and industrial sites, where residents had expressed concerns about their groundwater quality (**Figure 1.2**).

The POU treatment device selected for testing in this study, consisting of a single-stage, extruded activated carbon block, was installed underneath the kitchen sink at each of the participating residences and evaluated through monthly water quality testing for eight months. Influent and effluent water samples were tested for lead and other heavy metals (Chapter 2), PFASs (Chapter 3), and microbial indicator organisms (Chapter 4) each month. Additionally, study participants filled out a questionnaire at the beginning and at the end of the study to evaluate their perceptions of their water quality and of POU water treatment as an intervention, including perceived benefits, barriers, and self-efficacy (Chapter 5).

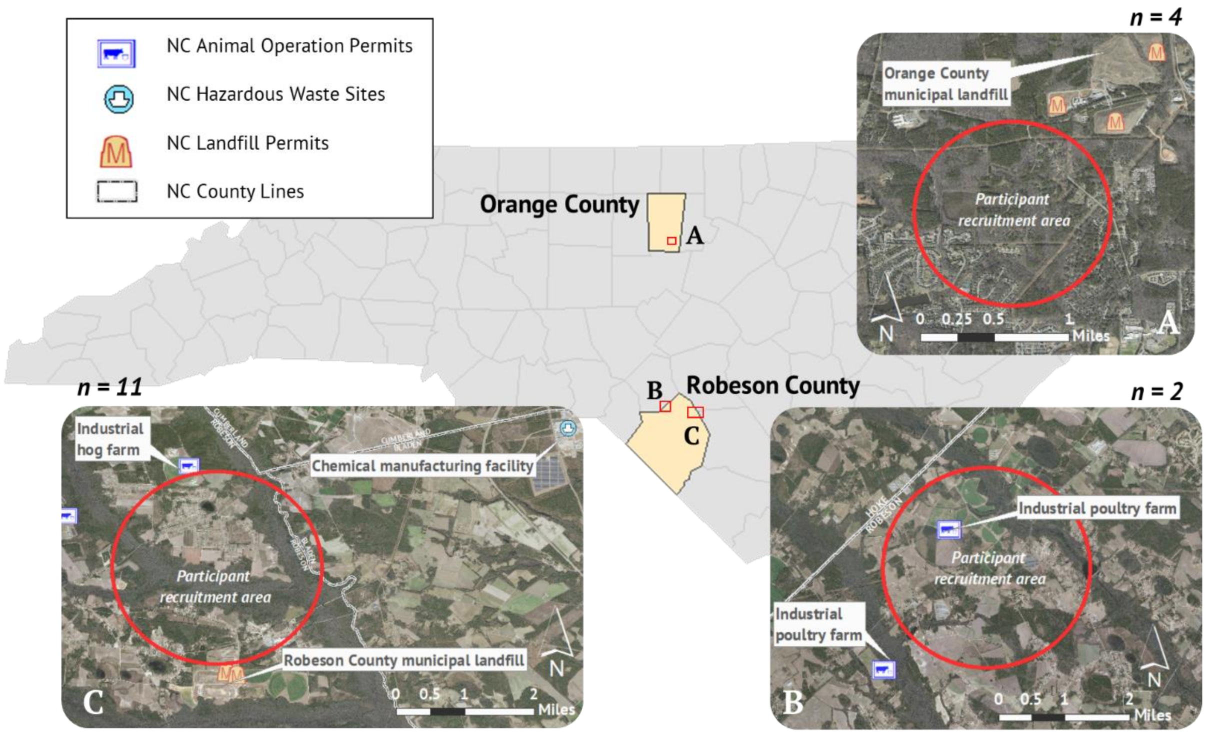

Figure 1.2. Map of study participant recruitment areas near potential environmental hazards in Orange county and Robeson County, North Carolina.

1.3.2 <u>A Solutions-Oriented Research Approach</u>

This work follows an interdisciplinary and solution-oriented research paradigm with immediate relevance for specific actions to protect public health as opposed to the dominant reductionist paradigms in environmental engineering science and environmental health research focused on advancing the scientific community's understanding of basic mechanisms of environmental processes, in both natural and engineered systems, and etiologic pathways of disease and risk (see Robinson and Sirard, 2005).

As Robinson and Sirard argue, the latter problem-oriented approach in biomedical and epidemiological research generally focuses on causes of past problems, which may inform hypotheses for solutions but ultimately leaves many important applied research questions—such testing the effectiveness of new policy or practice interventions—unanswered. Indeed, far more attention is paid in the scientific literature to the presence, causes, and consequences of contaminants in private wells than to testing practical interventions for addressing such exposures. Additionally, the reductionistic, problem-oriented approach in engineering tends to circumscribe innovation to designing and testing cutting-edge technologies in laboratory settings or theoretical advancements that have little relevance to immediate realities of everyday exposures and health risks. Solving the public health challenge of private well water requires a holistic, iterative, and deliberately practical approach that tests hypotheses and interventions in real-world settings. This research provides an example of such an approach within environmental engineering that may be replicated to continue to advance public health practice around private well water management and to benefit communities experiencing water quality risks in other contexts.

1.4 Dissertation Structure

This dissertation is structured in a nontraditional format as four separate manuscripts for publication in scholarly journals. As such, some information and references are repeated in multiple sections as each chapter is intended to stand alone for peer-review. The structure of each chapter also varies slightly according to specific journal guidelines. Chapter 2 originally appeared as an article in the journal *Water*.

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CHAPTER 2: UNDER-SINK ACTIVATED CARBON WATER FILTERS EFFECTIVELY REMOVE LEAD FROM PRIVATE WELL WATER FOR OVER SIX MONTHS[1]

2.1 Introduction

Private well users in the United States (U.S.) may be at elevated risk of exposure to lead (Pb) in drinking water than populations connected to community water systems (Macdonald Gibson et al., 2020). Pb can leach from borehole and household plumbing components where groundwater is corrosive (Pieper et al., 2015b, 2018), and is known to interfere with neurological development in children, even at low levels of exposure (ATSDR, 2007; Canfield et al., 2003; Lanphear et al., 2005). However, due to the fact that private wells are not regulated by the Safe Drinking Water Act (SDWA), elevated Pb levels and other drinking water contaminants in private well water often go unnoticed (Fizer et al., 2018; Stillo et al., 2019).

In response, a body of research has called for wider implementation of protections for well users such as increased risk communication to promote well testing (Colley et al., 2019; Flanagan et al., 2018; Morris et al., 2016; Stillo et al., 2019; Wood, 2019) and removing financial barriers to implement household and point-of-use (POU) water treatment for Pb (Macdonald Gibson et al., 2020; Nigra, 2020). However, even where increased well testing is achieved, knowledge gaps exist around the effectiveness of POU water treatment devices for well users since few U.S. studies have characterized the effectiveness of consumer water filter

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performance in household settings under real-world conditions over extended periods of time. In a systematic review of 3,142 POU drinking water filtration papers, Brown et al. (2017) found only 15 studies addressing POU filter effectiveness for chemical contaminants in the U.S. or Canada, only one of which reported data on Pb in private well water. This latter study surveyed 31 households in rural Arizona obtaining their water from a variety of sources (community systems and private wells) about their use of water treatment (Lothrop et al., 2015). Among the participating homes, 13 had a water treatment system of some type (water softener, reverse osmosis system, or activated carbon (AC) filter), eight of which were connected to private wells. Of the 13 homes using water treatment, nine were tested for Pb removal effectiveness, although the authors did not report which of these homes used private well water. Among these nine households, one-time sampling found that a water softener removed 71% of influent Pb; reverse osmosis systems (n=4) removed 61–90% of influent Pb; and AC systems (n=4) showed inconsistent performance. Pb removal in the four AC filters tested ranged from 31% to -16% (i.e., the Pb concentration was 16% higher after the filter than before in one home), but influent Pb levels for these homes were not reported. In a later study, not included in the review by Brown et al., Tomlinson et al. (2019) tested pour-through AC filters for Pb removal from well water in two households in North Carolina. In this study, 99% of first-draw Pb was removed at the time the filters were distributed, with influent Pb levels ranging 21–66 μg/L, but filters were not tested again thereafter.

These studies have very limited use in understanding the in-situ performance of POU water filters for Pb removal in homes relying on private wells. First, the cross-sectional design of both studies does not allow for changes in filter performance over time to be evaluated. Lothrop et al. (2017) collected only one-time samples and had no way of evaluating the cumulative volume of water treated by each device at the time samples were taken, while Tomlinson et al. (2019) only tested the filter effectiveness at start-up. Second, these studies could not analyze the removal effectiveness with respect to other influent water quality parameters or usage patterns, which vary greatly among households, given their limited sample sizes (n=9 and n=2, respectively). Third, Lothrop et al. (2015) only considered the effectiveness of water treatment systems already in place and did not report whether the devices were certified for Pb removal according to standards for household water treatment products put in place by the National Sanitation Foundation (NSF) and the American National Standards Institute (ANSI). Finally, in the study by Lothrop et al. (2015), water samples were collected from household taps after a two-minute flush in contrast to USEPA protocols for Pb sampling which require first-draw samples from taps after at least a six-hour stagnation period to represent worst-case exposure conditions (USEPA, 2002).

As a result, these previous studies provide little actionable information for well users or for state agency personnel charged with providing technical advice to well users. To date, the only longitudinal evaluations of POU filter effectiveness for metals in private well water in the U.S. have been tailored to arsenic removal (Möller et al., 2009; Powers et al., 2019; Spayd et al., 2015). What is more, despite rigorous certification standards under NSF/ANSI Pb reduction claims require systems to be tested using highly treated water adjusted to precise ranges for pH, alkalinity, and hardness which are not characteristic of many raw groundwaters (NSF Joint Committee on Drinking Water Treatment Units, 2018). Water treatment processes verified under precisely controlled conditions in laboratory settings or on municipally treated drinking water cannot be assumed to behave the same when applied in novel contexts, such as private wells.

Thus, there is a critical gap in current literature leaving millions of well users without evidence-based information for protecting against Pb in their water. To fill these gaps, this study provides the first longitudinal evaluation of POU water filters to remove Pb from private well water as a function of multiple in situ variables, including time in operation, volume of water treated, usage patterns, influent water quality, and Pb sources. Conducting solutions-focused research centered on improving decision-making around currently available technologies for private well users is both innovative and necessary toward improving environmental health in rural communities. The principal objective was to relate POU filter performance to household water usage, water quality characteristics, and Pb sources in a sample of real-world users to evaluate the range of performance that can be expected and inform individual well users, public health and well water professionals, and policymakers alike.

2.2 Materials and Methods

2.2.1 Recruitment of study participants and baseline Pb levels

Households served by private wells were recruited in three geographic clusters (A, B, and C) in Orange County and Robeson County, North Carolina (**Figure A.1**). Cluster A is majority white and middle-income while clusters B and C are racially diverse and predominantly low-income (**Table A.1**). These areas were selected through the help of community partner organizations that had identified areas of suspected groundwater contamination. Participants were recruited by e-mail, flyers, word-of-mouth, and door-to-door invitations with a community partner. Twenty households were initially recruited to participate in baseline testing consisting of a 250 mL first-draw sample (6-hour stagnation time) collected at the primary kitchen faucet. The U.S. Environmental Protection Agency (USEPA) Lead and Copper Rule requires one liter first-draw samples in regulated community water systems, but a 250 mL first draw was used for comparison with previous studies on Pb in private well water (Pieper et al., 2015b, 2015a). Certified pre-cleaned, wide-mouth HDPE bottles were delivered to participating households the day before sampling and instructed to collect the first-draw water (i.e., water collected from the faucet after a minimum six-hour stagnation time without prior flushing) from the kitchen tap in the morning. Samples were transported to the University of North Carolina at Chapel Hill (UNC) and transferred to 10 mL aliquots, acidified to 2% nitric acid, and stored at 4° C before analysis. Analytical methods are discussed in Section 2.2.5 .

The mean first-draw Pb concentration among the 20 households invited to participate was 9.3 µg/L (median=8.2 µg/L) and ranged from 0.1 to 34.3 µg/L. Three households (15%) exceeded the USEPA's action level of 15 µg/L and sixteen households (80%) had 250 mL first-draw Pb concentrations above the American Academy of Pediatrics' recommendation of 1 µg/L for water fountains in schools (American Academy of Pediatrics Council on Environmental Health, 2016). Previous testing at the kitchen tap using 250 mL first-draw samples in North Carolina (n=14) and Virginia (n=2,144) has shown a similar prevalence of Pb occurrence, with 14 – 19% of wells having first-draw Pb levels above 15 µg/L and 82 – 93% of wells having first-draw Pb above 1 µg/L (Pieper et al., 2015a, 2015b), suggesting that the first-draw Pb levels seen in this study are comparable to levels across in the region.

After baseline testing, 17 households opted to receive the water filter for Pb testing. Of these, two households relied on the same well. Six (35%) were built prior to 1986 (i.e., when the SDWA was amended to ban pure lead plumbing and limit household components to <8% Pb by weight (*Safe Drinking Water Act Amendments of 1986*, 1986)), and 16 (94%) were built prior to 2014 (i.e., when the SDWA was amended to further limit plumbing components to <0.25% Pb in wetted surfaces (*Reduction of Lead in Drinking Water Act*, 2011)) (**Table A.2**). This study was approved by the UNC Institutional Review Board (study number 19-1015).

2.2.2 POU intervention design

The selection criteria for the POU device in this study included: a full-flow, under-sink design; activated carbon (AC)-based; certified to NSF/ANSI 53 for Pb reduction, NSF/ANSI 42 for particulate reduction, and NSF P473 for per- and polyfluoroalkyl substance (PFAS) removal (methods and results for PFAS removal are forthcoming); and widely available. An AC device was chosen over a reverse osmosis system since AC filters represent lower capital, operation and maintenance costs (USEPA, 2007), and generate significantly less waste and utilize less water compared to reverse osmosis (USEPA, 2006). Reverse osmosis membranes may also negatively affect the aesthetic quality of drinking water (Thomas et al., 2019) and can degrade rapidly (Pratson et al., 2009). Thus, in the long-term, AC-based filters may be a more economical, user-friendly, and sustainable household treatment solution for well users addressing Pb. The selected device is distributed by national hardware stores and costs $100 initially and $70 for each replacement filter cartridge. The manufacturer-recommended lifetime of the cartridge is six months for a rated capacity of 2,967 L, representing a maintenance cost of approximately $12 per month if the cartridge is replaced at the recommended interval.

Filters were installed beneath the primary kitchen sink at each participating household (**Figure 2.1**). The device treats the full flow of cold water at the main faucet with a rated flow rate of up to 5.67 L/min. The filter is comprised of a single-stage, extruded solid AC block. According to the manufacturer, the block is produced using a coconut-shell powdered AC mixed with a metals scavenger material – possibly silicon/titanium oxides, as documented elsewhere (Deshommes et al., 2012) – and a proprietary binding agent (2019, personal communication, 24

September). The filter does not include any prefilter fabric or membrane around the surface of the block. To evaluate the cumulative volume of water treated over time, a food-grade polypropylene flow sensor (Sea YF-S201 or Gredia GR-301) and a data logger (Onset Hobo State Logger) were installed in-line with each system. Loggers were set to record at 10 second intervals to capture detailed water usage patterns. Pb-free polypropylene sample ports were installed at the filter influent and effluent underneath the sink. All tubing used in the system was made of food-grade polyethylene.

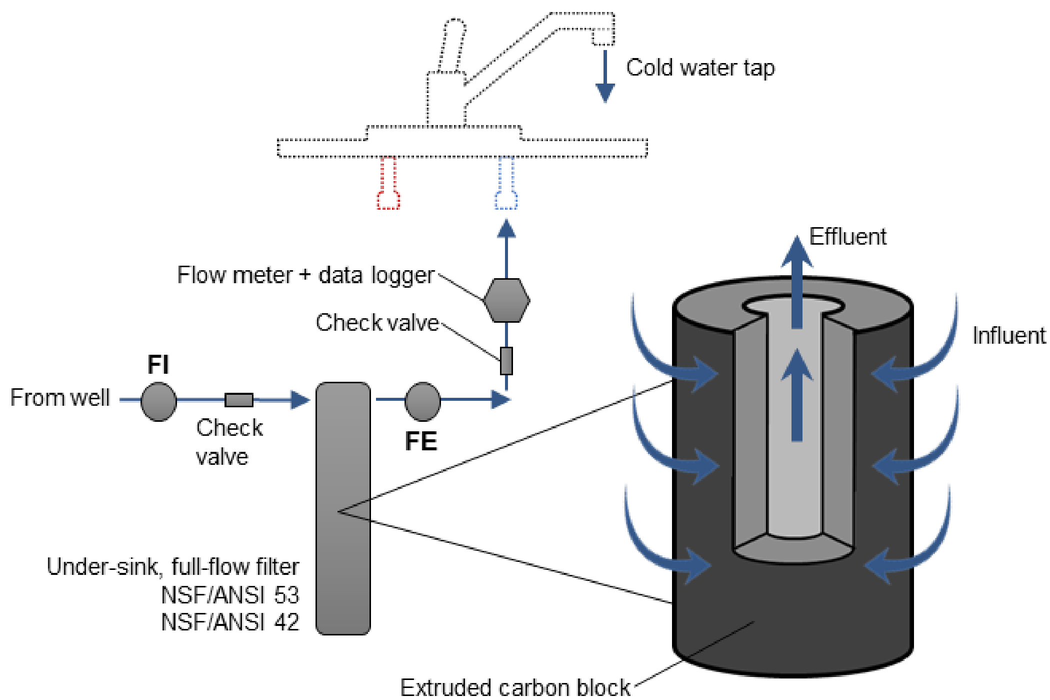

Figure 2.1. Schematic of filter installation. FI = "Filter Influent." FE = "Filter Effluent."

2.2.3 Influent groundwater quality

Clusters B (n=2 households) and C (n=11 households) had aggressive water with average (± SD) pH levels of 5.43 (± 1.13) and 4.43 (± 0.26), respectively (**Table A.3**). The average Langelier Saturation Index (LSI) and chloride-to-sulfate mass ratio (CSMR) indicated very high corrosion risk in both clusters (LSI <-0.5, CSMR >1). These conditions are representative of many groundwaters across the southeastern U.S. (Belitz et al., 2016). Waters in cluster A (n=4 households) had an average pH of 7.07 (± 0.43) and exhibited lower corrosion risk due to greater hardness and carbonate alkalinity. Influent turbidity and dissolved organic carbon in all wells were generally low (< 1 NTU and <1 mg/L, respectively). Overall, the average influent water quality in each cluster was outside the range required for certification of Pb removal under NSF/ANSI 53 with respect to pH, hardness, or alkalinity.

2.2.4 Sampling methods

2.2.4.1 Monthly sampling

After filter installation, influent and effluent samples were collected monthly for approximately eight months from October 2019 to June 2020. Two sampling months in the middle of the study (April – May 2020) were lost due COVID-19 restrictions. Samples were collected in virgin, one-liter, acid-washed, wide-mouthed HDPE bottles. Bottles were soaked in 3 M nitric acid solution for a minimum of three days then rinsed with deionized water five times prior to sample collection. Previous POU filter assessments for Pb have collected influent and effluent samples sequentially on the same day, but have recognized the limitations of this approach for calculating removal given the non-constant nature of Pb in premise plumbing (Bosscher et al., 2019; Deshommes et al., 2012). To account for this, study participants were trained to collect first-draw samples at labeled influent and effluent sample ports beneath their kitchen sink on two consecutive days. This protocol ensured that calculations of the filter's removal effectiveness were based on first-draw conditions at both the influent and effluent. If study participants neglected to fill their sample bottles, random daytime samples were taken from each sample port at the time of the researcher's visit which have been shown to adequately estimate first-draw Pb levels (Riblet et al., 2019). In these instances, the influent was sampled before the effluent. Sample bottles were collected from participating households each month and transported on ice to UNC where they were transferred into 10 mL aliquots and acidified to 2% nitric acid (Plasma Pure®, SPC Science, Montreal, CA). Acidified samples were stored at 4° C before analysis.

Additional samples were also collected from the influent and the effluent at the time of each visit to evaluate changes in microbial water quality across the filters (complete microbial methods and results will be reported in a forthcoming manuscript).

2.2.4.2 Pb profiling

Five households were selected for sequential sampling from the main faucet before and after the filter was installed to profile the occurrence of Pb within the household plumbing. A standardized sampling protocol was adapted from Pieper et al. (2015) that could be easily implemented by study participants (Pieper et al., 2015a). The protocol (**Figure 2.2**) entailed a 250 mL first-draw sample, immediately followed by three consecutive one-liter samples without any flushing in between. The faucet was then flushed for one minute and five minutes at full flow with the fifth and sixth one-liter samples filled after each flushing interval. Pieper et al. (2015a) have shown that, in households connected to private wells, the volume between the kitchen faucet and pressure tank is typically ≤ 3 liters. Thus, samples 1-4 in the sequence approximate the profile of water between the faucet and the pressure tank, while samples 5 and 6 represent water from the pressure tank and borehole components. This simplified protocol was used as a rapid screening tool that allowed for a) detection of general sources of Pb (e.g., from the faucet and sink fittings alone or from elsewhere in the system), and b) evaluation of the effectiveness of faucet flushing with and without the filter in place. All sample bottles were either certified pre-cleaned or acid washed, as above. Additional 10 mL aliquots were drawn from a subset of samples and passed through a 0.45 µm nylon syringe filter (GE Whatman

GD/XP) on the same day. Both the filtered and unfiltered 10 mL aliquots were then acidified as above and stored at 4°C before analysis. The filtered samples characterized dissolved Pb, while the difference between filtered and unfiltered samples was calculated to estimate particulate Pb (Pieper et al., 2015a).

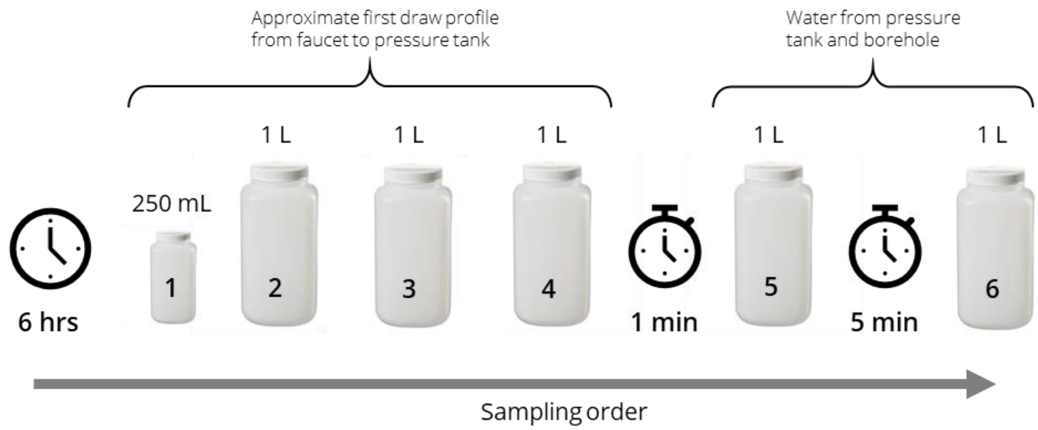

Figure 2.2. Volumes and flushing times for sequential sampling before and after filters were installed in five households with elevated first-draw lead concentrations.

2.2.5 Analytical methods

Samples were analyzed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) adapted from USEPA Method 6020B (USEPA, 2014) on an Agilent 7500cx instrument. Acidified samples were centrifuged at 5,000 RPM for 8-10 minutes before analysis to separate any suspended solids in solution. In addition to Pb, samples were analyzed for corrosion byproducts and other metals including aluminum (Al), manganese (Mn), iron (Fe), copper (Cu), nickel (Ni), zinc (Zn), arsenic (As), cadmium (Cd), tin (Sn), and uranium (U). A combined ten-point calibration curve was prepared for all elements before each sample run. The full ICP-MS instrument configuration and operation conditions have been described previously (Tomlinson et al., 2019). Per USEPA guidelines, the limit of quantitation (LOQ) was determined as ten times the standard deviation of the replicate blanks, or 0.015 µg/L for Pb, and non-detect results were assigned a value of one-half the LOQ (USEPA, 1991).

Quality control measures for metals quantification included laboratory blanks; field blanks; replicate analyses performed every 10 samples; and verification of instrument performance using a National Institute of Standards and Technology certified reference material (CRM) for trace metals in drinking water (High Purity Standards, Charleston, SC.) Sn is not included in the CRM and was spiked in at known concentrations. The mean recovery for all metals in the CRM, including Sn, was 102%. The relative standard deviation of all repeat measurements was ≤10% and the average difference of all sample replicates was 7.6%.

Field measurements of temperature, pH, and electrical conductivity were taken using a handheld probe (HI98129, Hanna Instruments, Smithfield, RI) calibrated with a two-point calibration in the field each day before use.

2.2.6 Data analysis

Paired influent and effluent samples were evaluated for statistically significant reductions of each metal at each sample month using non-parametric Wilcoxon signed rank tests for non-normally distributed samples. The appropriateness of the Wilcoxon method was evaluated by the Shapiro-Wilk test. Reported *p*-values for filter performance represent the results of two-sided paired Wilcoxon tests unless otherwise noted. Additionally, the American Academy of Pediatrics' recommendation of 1 µg/L of Pb in drinking water was used to evaluate the filters' protectiveness as a conservative health-based goal (American Academy of Pediatrics Council on Environmental Health, 2016). Although the USEPA Lead and Copper Rule action level of 15 µg/L is an established regulatory threshold, the action level is designed to be used as a utility-scale indicator of the effectiveness of corrosion control in drinking water distribution systems rather than as a measure of individual health risk (Triantafyllidou and Edwards, 2012). Indeed, the action level has been shown to be an unsafe level in drinking water for the most vulnerable population groups and the USEPA has set a maximum contaminant level goal of no lead in drinking water (Redmon et al., 2018). Furthermore, households connected to private wells are not included under the Lead and Copper Rule or SDWA stipulations. Thus, we considered the American Academy of Pediatrics' recommendation to be a more appropriate threshold for evaluating health risk of lead in drinking water, especially for children.

2.3 Results and Discussion

2.3.1 Long-term filter effectiveness for Pb removal

Filters decreased the influent Pb to below the American Academy of Pediatrics' recommendation of 1 µg/L in all 17 households for the entire duration of use (**Figure 2.3**). In three households (18%) filters had to be removed within 2–3 months due to clogging (see Section 2.3.2), but the filters remained operational in the remaining fourteen houses for the full eight-month study duration. Paired influent-effluent samples exhibited a highly statistically significant reduction in Pb across all sample points ($p < 0.0001$; **Figure 2.4**). Excluding 10% of paired samples where participant sampling error was suspected, the mean removal among all samples and all households was 97.8%. Importantly, Pb removal was consistent across all households and geographic clusters, indicating that the filter's Pb removal effectiveness was independent of both the influent groundwater quality and the variations in water usage patterns observed in this study.

This study is unique in that no other longitudinal assessments of POU filters for Pb removal from private well water are currently available. As discussed previously, other studies have evaluated POU effectiveness for Pb removal from private well water through limited cross-sectional sampling. In two households in North Carolina, Tomlinson et al. (2019) showed that pour-through AC filters removed 99% of first-draw Pb at the time the filters were distributed, with influent Pb levels ranging 21 – 66 μg/L (Tomlinson et al., 2019). Pour-through devices are low-cost and easily implementable, but Deshommes et al. (2010) have observed that these devices exhibit worse performance over time than under-sink and faucet-mounted devices due to short-circuiting through loose granular media and poor removal of particulate Pb, putting into question the long-term protectiveness of pour-through devices for well users (Deshommes et al., 2010). In a survey of four homes using various faucet-mounted or under-sink AC devices relying on both community water systems and private wells in rural Arizona, Lothrop et al. (2015) found that Pb removal ranged from -16% to 37%, although the authors did not report clearly which samples were from private wells (Lothrop et al., 2015). The difference between the results of Lothrop et al. and those shown here may be related to the certification of each device and/or the length of time it had been in use and indicate that deteriorating performance and possible desorption of previously retained Pb may occur in certain AC devices over time.

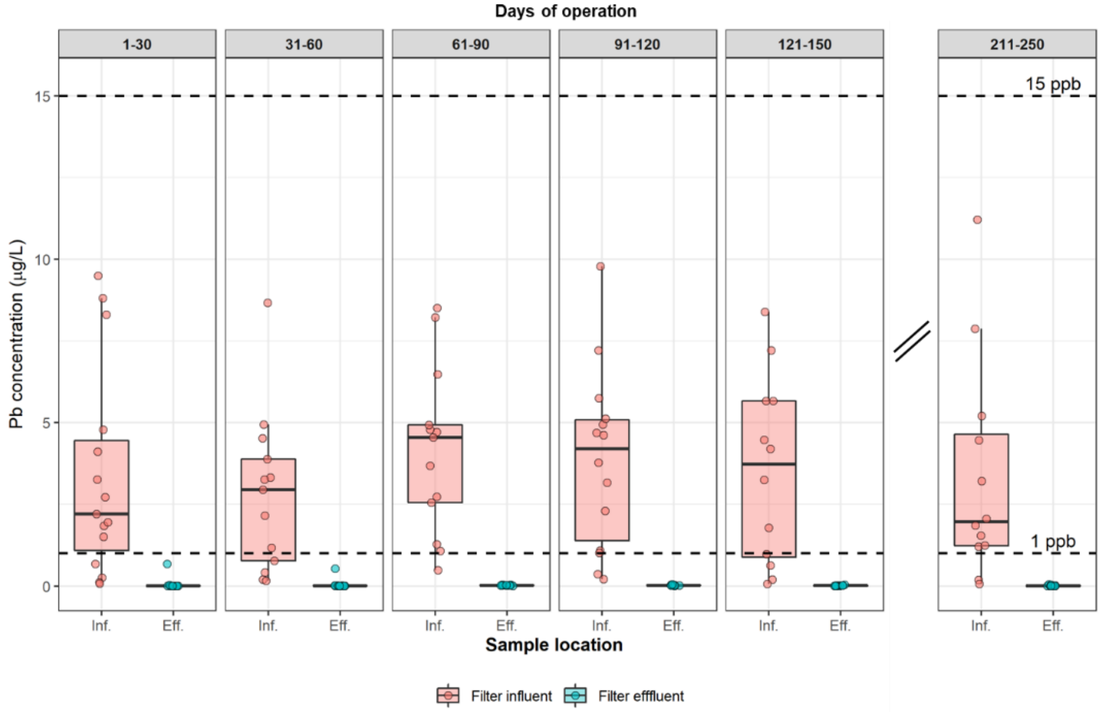

Figure 2.3. Distribution of influent and effluent Pb concentrations among all study households at each sampling time. Vertical panes show the number of days of filter operation for each collection of samples. Filter influent samples (Inf.) shown in pink. Filter effluent samples (Eff.) shown in teal.

Although not from private well water, several studies of POU devices installed in situ with municipal tap water provide useful comparisons. Most recently, 97% of effluent samples from 345 faucet-mounted filters installed in Flint, Michigan during the water crisis were below 0.5 µg/L (Bosscher et al., 2019), compared to 95% of effluent samples below 0.5 µg/L in this study. Additionally, Deshommes et al. (2012) tested the effectiveness of five under-sink NSF/ANSI 53-certified AC devices for Pb removal in a large building connected to a municipal water supply and found effective removal over one year, with median influent Pb levels of 111 µg/L reduced to a maximum of 2.2 µg/L (Deshommes et al., 2012). Similarly, Boyd et al. (2005) demonstrated that 17 under-sink filters installed at drinking water fountains in schools also reduced influent Pb levels ranging 1 – 93 µg/L to <1 µg/L during accelerated testing over the course of one month, although three of the 17 filters clogged prematurely (Boyd et al., 2005). In general, these studies support the results shown here and indicate that under-sink AC filters deployed for private well waters can achieve comparable Pb removal as municipal tap water, although pre-treatment may be necessary to prevent clogging in both scenarios.

2.3.2 Filter failure due to clogging

The rate of premature failure due to clogging observed by Boyd et al. (2005) was the same as what was observed in this study (approximately 18%) (Boyd et al., 2005). In both studies, Pb was still effectively removed even at terminal flow conditions. Boyd et. al. identified the main cause of clogging as high Fe concentrations (up to 28 mg/L) in the influent from corrosion of galvanized steel pipes in some schools. Clogging occurred after treating only 30 – 40% of the filters' rated capacity. High Fe was also the most likely cause of clogging in one household in this study (#21) where influent concentrations exceeded 4.5 mg/L. Influent Fe concentrations in the other two clogged filters were low (<0.1 mg/L). In one of these households (#16), clogging appeared to be due to extremely low use during the first month after installation, possibly allowing rapid biofouling of the carbon (results on microbial growth with the filters are forthcoming), while the cause in the third household (#17) was not apparent. Given that all filters operated normally at start-up, clogging was presumed not to be the result of a faulty device. Clogging occurred after 2-3 months of use, representing 150 – 1,335 L of water treated (5 – 45% of the rated capacity). All other filters remained usable for the duration of the study although the maximum daily flow rate was generally low (2.2 L/min on average, sd=0.83; **Figure A.2**). A sediment pre-filter to remove high influent Fe and/or intermittent turbidity from private well water may be necessary to reduce clogging and extend the filter's life.

2.3.3 Removal of other metals

The filters also achieved highly statistically significant reductions ($p < 0.0001$) in the median effluent concentrations of Al, As, Cd, Cu, Fe, Sn, U, and Zn over the study duration (**Figure 2.4**, **Table A.4**). Significant reduction of Ni ($p < 0.01$) was also observed, but median Ni effluent concentrations began to approach and even exceed the influent concentrations after four months of use, indicating that more highly adsorbing metals may displace previously adsorbed Ni (**Figure A.3**). This phenomenon has previously been observed for Ni (Deshommes et al., 2010) as well as Cd and Zn in lab-tested AC systems (Taylor and Kuennen, 1994). Mn was significantly reduced in the first month of testing ($p < 0.001$), but quickly achieved breakthrough in subsequent months and was the only metal without a significant reduction in the median concentration when data were aggregated across all months.

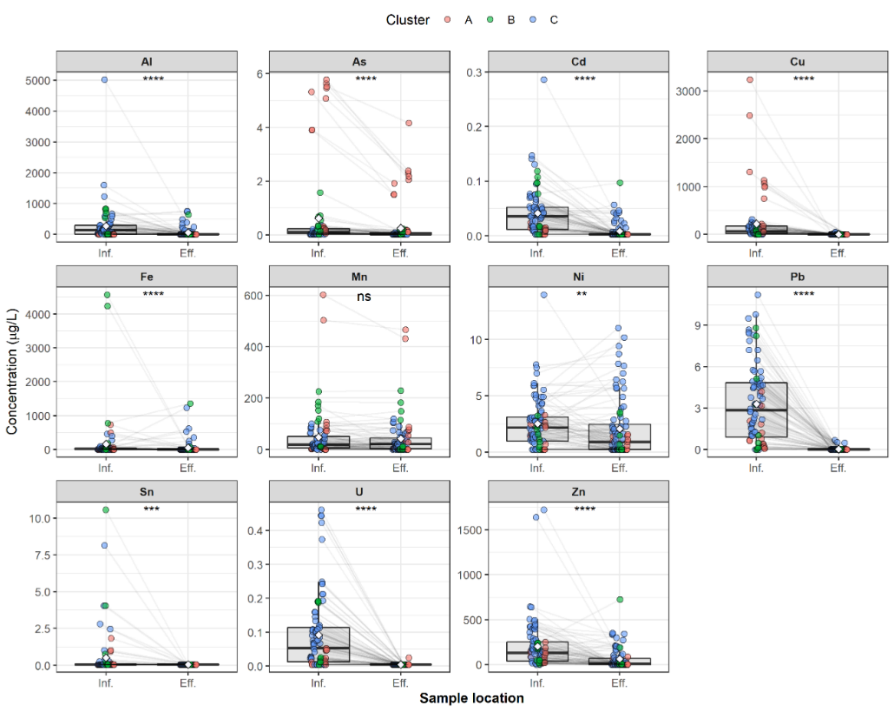

Figure 2.4. Paired influent and effluent samples across the filters aggregated from all households and sample months for each metal analyzed. The color of each point indicates the geographic cluster of the household: A (red), B (green), and C (blue). Stars indicate significance of difference between influent and effluent concentrations in one-sided Wilcoxon tests. ns: $p > 0.05$; ** $p < 0.01$; *** $p < 0.001$; **** $p < 0.0001$.

2.3.4 Contribution of the faucet to first-draw Pb

The Pb levels in the filter influent (i.e., collected from sample ports beneath the kitchen sink; **Figure 2.1**) were significantly lower than the first-draw Pb concentrations in baseline samples collected from the faucet fixture itself ($p < 0.005$; **Figure 2.5**, **Table A.5**). Without the filter, the Pb concentration in the initial 250 mL first-draw sample at baseline averaged 9.0 µg/L (sd=10.1, maximum=34.3 µg/L). In comparison, samples from the filter influent (indicating water quality before interaction with the tap) averaged 3.3 µg/L (sd=2.4, maximum=8.4 µg/L). This difference implicates the faucet fixtures as an important Pb source, as documented elsewhere (Pieper et al., 2015a, 2018). The faucets in all but one home (#13) were installed prior to more stringent Pb-composition standards were put in place in 2014 (*Reduction of Lead in Drinking Water Act*, 2011), but even the faucet installed after 2014 had 77% more Pb in the baseline first-draw sample than the average filter influent. Indeed, the NSF/ANSI standard that evaluates Pb-leaching from plumbing components does not require testing under highly corrosive conditions and, thus, “Pb-free” components may still leach significant Pb under conditions commonly seen in private wells (Lei et al., 2018; Pieper et al., 2016). Cd, Cu, and Zn were also found in higher concentrations in the first 250 mL of the profiles compared to the flushed water, indicating clear corrosion of the faucet components consistent with prior studies (**Figure A.4**) (Samuels and Méranger, 1984). In one household (#2), the 250 mL first-draw Cd concentration without the filter was consistently three to four times the USEPA Maximum Contaminant Level Goal of 5 µg/L (16.4 µg/L during profile sampling and 22.1 µg/L during baseline sampling). All other first-draw Cd concentrations were below 1 µg/L. In addition, Spearman’s correlation coefficients showed that Pb was strongly correlated with Al (β= 0.59, $p <$ 0.05), Cd (β= 0.74, $p < 0.001$), and Zn ($\beta = 0.64$, $p < 0.01$), but not with Cu ($\beta = 0.24$, $p = 0.22$)

in the baseline first-draw samples, suggesting that impurities in die-cast zinc-aluminum alloy (a material known as Zamak typically used in low-cost and internationally manufactured faucets) rather than brass faucet components may be a contributor of Pb and Cd in some homes (Otunniyi and Oluokun, 2014; Sheppard, 2020; StarCraft Custom Builders, 2018).

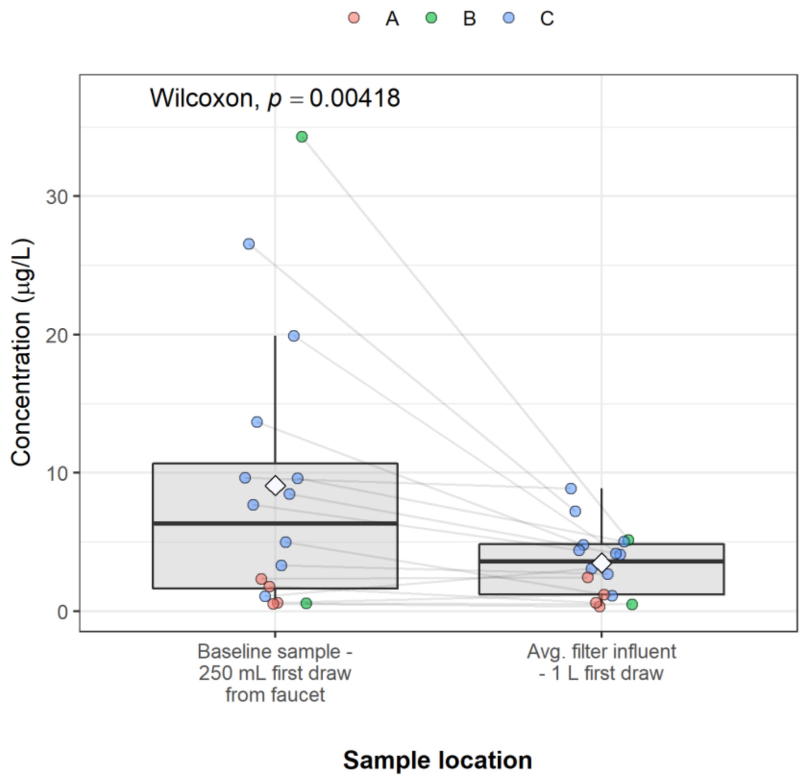

Figure 2.5. Comparison between baseline first-draw Pb levels in samples collected directly at the kitchen faucet (left) and average filter influent Pb levels in samples collected beneath the kitchen sink from a sample port without interaction with the faucet (right). Colors show geographic cluster of the household and lines show paired household samples. White diamonds show the group mean.

2.3.5 Pb profiling results

Profile sampling was conducted in five of the highest-risk homes in clusters B and C before the filter was installed and again after three months of use (**Figure 2.6**). In one home (#17 in **Figure 2.6**), the filter clogged after just three months of use before the second round of profiling could be completed.

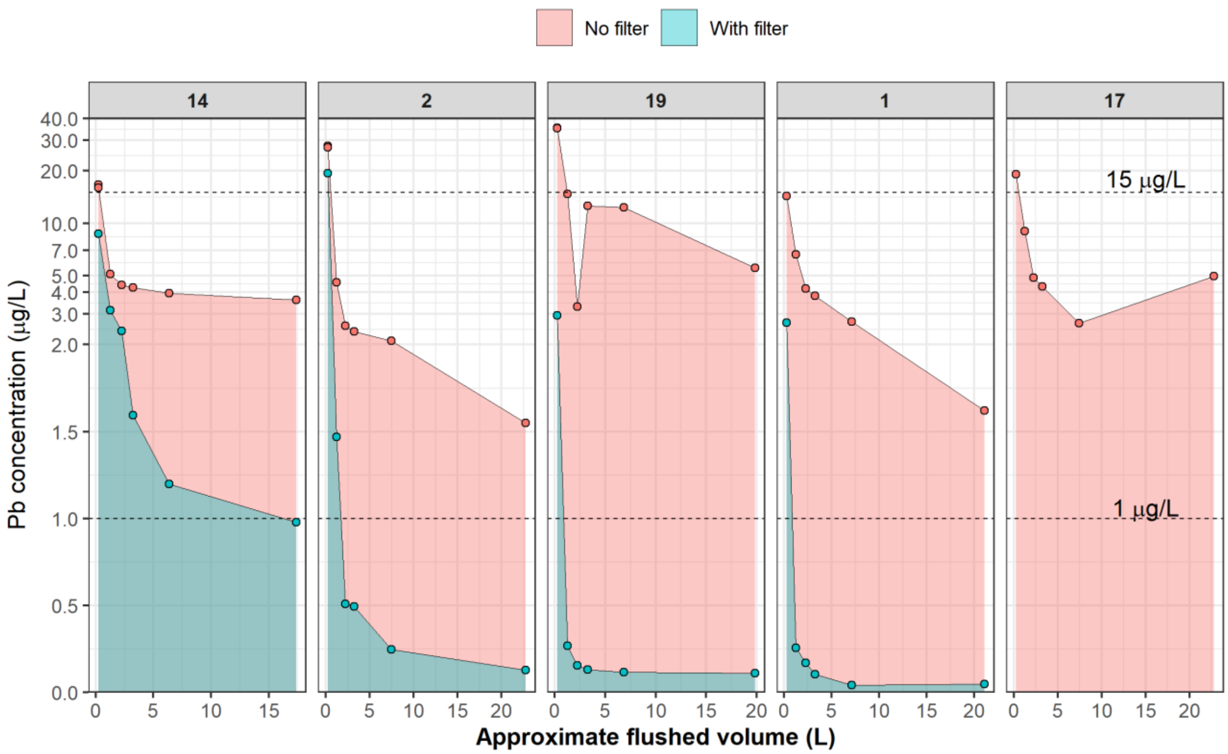

Figure 2.6. Results of Pb profile sampling in five households before and after filter installation.

2.3.5.1 Flushing effectiveness without filter

Without the filter installed, the Pb concentration decreased with flushing but remained above the American Academy of Pediatrics' recommendation of 1 µg/L even after approximately eight minutes (**Figure 2.6**, pink profiles). In two homes (#19 and #17), Pb initially decreased with flushing, then increased again. Over 98% of the Pb in these spikes was in dissolved form, indicating leaching from solder, brass, or galvanized steel pipe (Clark et al., 2015; Pieper et al., 2015b) rather than scouring of particulate Pb-bearing scale as the source. Thus, these households represent a different type of Pb release than previously characterized by private well water profile sampling, where semi-random increases in Pb concentrations in the profile were predominantly in the particulate form (Pieper et al., 2015a). Otherwise, this finding confirms what has been shown elsewhere that flushing can reduce Pb levels at the tap, but not ensure that the water is consistently safe for consumption (Katner et al., 2018; Pieper et al., 2019).

2.3.5.2 Improved flushing effectiveness with under-sink filters

With the filter installed, the Pb profiles showed rapid and consistent decreases in the Pb concentration. In three households, the Pb concentration decreased to less than 1 µg/L within two liters of flushing (generally one minute or less). In household 14, the rate of decrease was lower, possibly indicating greater leaching from the faucet components. In general, the post-filter profiles demonstrate that the filter effectively decreased Pb levels at the tap but that water can still be contaminated by the fixture after treatment. This vulnerability of under-sink filters has also been observed in buildings connected to municipal waters (Boyd et al., 2008a, 2008b; Deshommes et al., 2012).

Even so, the filter also decreased the amount of Pb in the first-draw water. On average, the concentration of Pb in the 250 mL first-draw sample was 64% lower with the filter installed, presumably as a result of dilution with filtered water and increased pH in the filter effluent (see Section 2.3.6.2). Furthermore, the filter improved the effectiveness of flushing itself. On average, flushing 2.25 L (approximately one minute) reduced Pb levels at the tap by 93% with the filter compared to a reduction of only 76% without the filter, indicating a 22% increase in the effectiveness of flushing. The Pb concentration in the approximate one-minute flush water was also reduced by 85% on average when the filter was installed (mean Pb without a filter = 5.5 µg/L; mean Pb with the filter = 0.6 µg/L). On a mass basis, the total Pb mass in profiles without the filter was approximately 49.1 – 155 µg compared to 2.1 – 23.9 µg with the filters, representing a mass reduction of 66 – 98% (85% on average) in the first 17 – 23 L of flushed water (depending on flow rate) after a 6-hour stagnation time. Thus, even with additional risks due to the faucet, the use of an under-sink filter reduces the total amount of Pb at the tap by a) mitigating Pb release from distant plumbing sources, ensuring that users are not inadvertently exposed to higher concentrations of Pb in the flushed water (Katner et al., 2018), and b) improving the effectiveness of flushing by requiring less flushing time to reach Pb levels below 1 μg/L. Installing a filter in conjunction with flushing the faucet for one minute after long periods of non-use will thus ensure the greatest Pb exposure reduction.

2.3.6 Factors influencing filter performance

2.3.6.1 Water usage patterns and surface loading

The observed long-term effectiveness of the filters for Pb removal may be attributable in part to relatively low water usage and Pb loading at the kitchen tap. Usage patterns collected from the data loggers revealed that, on average, each filter was in use for only 1 – 20 minutes per day, indicating that water was not flowing for over 23 hours per day. Rather than increased stagnation time leading to Pb breakthrough as previously hypothesized (Deshommes et al., 2010), long periods of non-use may increase the time allowed for intraparticle diffusion of the sorbate and thus improve removal as dissolved Pb ions penetrate further into the micropores of the carbon structure (Kuennen et al., 1992). Indeed, a follow-up to a lab-based assessment of under-sink AC filters for Pb (Deshommes et al., 2010) found that, when deployed under real-world conditions, the effectiveness of the same device improved slightly to what was observed during non-stop flow testing under laboratory conditions (Deshommes et al., 2012). Although NSF/ANSI 53 does require off periods, filters are operated on a continuous cycle for 16 hours per day followed by an eight-hour rest so are unlikely to capture the effect of extended stagnation time during certification testing.

Consistent with the low water use time observed, the cumulative volume of water treated by each filter in the first six months of the study (excluding the three filters that clogged after 2-3 months, see Section 2.3.2) ranged from 151 – 3,481 L (representing approximately 160 – 3,700 bed volumes, or 5 – 117% of the filter's rated capacity), with an average water usage of 1,063 (±799) L (**Figure A.5**). Only one household exceeded the filter's capacity of 2,967 L after six months. The reasons for this wide variation in usage patterns are not known but could include factors such as family size, presence of pets, household water pressure, and myriad behavioral factors related to cooking, cleaning, drinking water, and perceptions of water quality. For example, several households had a prior aversion to their well water and continued to supplement their water supply with bottled water throughout the study. In addition, some participants reported adapting to intentionally use their hot water supply for cleaning and washing more often to prolong the filter's life.

The observed Pb surface loading, i.e., the mass of Pb adsorbed per mass of carbon, was also low compared to the certification requirements. NSF/ANSI 53 certification requires filters to be challenged with a constant influent of 150 µg/L (NSF Joint Committee on Drinking Water Treatment Units, 2018). Considering the rated capacity of 2,967 L, the total loading during certification thus exceeds 445 mg of Pb. Manufacturers can claim only 50% of the successfully tested capacity of a filter if a performance indication device is not included (as is the case with the filter tested here), so the actual capacity is potentially even greater. The mass of carbon can be estimated using the volume of the block (950 cm^3) and the bulk density of coconut shell AC (~0.5 g/cm^3) (Mulhern et al., 2017), yielding a mass of approximately 475 g of carbon. Thus, during certification, the Pb loading on the filter was approximately 0.93 mg Pb/g carbon.

By comparison, the overall surface loading in practice was estimated by multiplying the average influent Pb concentration by the total volume of water treated for each household. First, the average influent Pb among the participating households was 0.13 – 8.37 µg/L (**Table A.5**). Thus, at the influent concentrations and rates of water usage observed, the estimated Pb loading of the filters was 0.76 – 9.51 mg during the first six months of use, or approximately 0.002 – 0.02 mg Pb/g carbon, representing only 0.2 – 2.1% of the filter's certified Pb load. This finding demonstrates that the manufacturer's stated capacity of 2,967 L is likely to be protective for most homes based on the influent concentrations and the rate of water usage observed in practice. Barring extreme scenarios of Pb release, such as after disruptions to the system or in the presence of pure Pb components (Deshommes et al., 2010), under-sink filters, which treat water that does not interact with the faucet, may only be consistently challenged by relatively low Pb levels even in high risk households like those in clusters B and C. Further research is required to understand how Pb loading may vary among other private well users as well as municipal water users.

2.3.6.2 Influent groundwater pH

Bench-scale column testing has shown that pH levels below 6 dramatically reduce the effectiveness of AC for Pb adsorption because, at low pH, carbon adsorption sites are more likely to be positively charged, thus repelling positively charged Pb ions in solution (Kuennen et al., 1992; Taylor and Kuennen, 1994). As a result, research suggests that POU AC devices should only be used for Pb control within a pH range of 5.5 – 10 (Kuennen et al., 1992). However, in cluster C, where pH levels were consistently below 5, with a minimum recorded pH of 3.9 (**Table A.1**), 98.5% of influent Pb was still removed throughout the study. This may be due, in part, to the low surface loading of the filters discussed above, suggesting that even under suboptimal groundwater conditions the carbon use rate is such that significant breakthrough is not observed during the recommended cartridge lifetime.

Additionally, in the previous experiments (Taylor and Kuennen, 1994), the AC studied was acid-washed before testing to remove hydroxyl groups and thereby minimize Pb precipitation. However, Pb precipitation – either on the carbon surface or in the carbon pore liquid – is one of the dominant removal mechanisms in AC systems (Goel et al., 2005; Largitte et al., 2014). Rinsing the carbon with a base solution after acid washing also significantly improves Pb removal, highlighting the importance of hydroxyl functional groups on the carbon's surface toward Pb precipitation (Reed, 1995; Reed and Arunachalam, 1994). Although the specific activation process and pre-treatment of the AC in the filter tested in this study is not known, the pH increased significantly in the effluent samples, with a greater increase at early time points (median influent pH at start-up of 5.23 compared to a median effluent pH at start-up of 9.13, $p < 0.0005$) and a gradual equilibration between the influent and the effluent by the end of the study (median influent pH at study end of 4.67 compared to a median effluent pH at study end of 4.73, $p = 0.86$; **Figure A.6**), suggesting that hydroxyl groups on the carbon surface are gradually exhausted (Sontheimer et al., 1988). Furthermore, Pb is highly soluble in acidic, low-alkalinity waters (Jurgens et al., 2019) like those in cluster C. Characterization of particulate and dissolved Pb levels during profile sampling in a subset of homes in cluster C confirmed that 98% of influent Pb was in the dissolved form. Thus, the dominant removal mechanism in low pH waters appears to be through precipitation of influent dissolved Pb ions on the alkaline carbon surface or in the pore liquid. While low pH may reduce the removal capacity due to adsorption-specific processes (Kuennen et al., 1992; Taylor and Kuennen, 1994), it does not appear to negatively impact removal by AC where Pb precipitation can occur.

Further research is needed to know whether acidic influent waters pose a risk for precipitated Pb to re-dissolve and be released in the filter effluent as the Pb solubility within the filter changes. Reed and Arunachalam (1994) showed that decreasing column pH corresponded with increasing Pb in the effluent of granular activated columns for wastewater treatment (Reed and Arunachalam, 1994). This behavior was not observed in the present study after two months of testing beyond the manufacturer recommended filter life, but it could occur if the filter cartridge is not replaced at recommended intervals. Six months was protective for the sample of well users in this study. In the absence of precise flow data, monitoring of the effluent pH to detect when it reaches influent levels may provide a simple method of determining when the carbon block needs to be replaced, with opportunities for improvements in POU monitoring through the use of smart technologies and remote water quality monitoring (Geetha and Gouthami, 2016; Hoffman et al., 2019).

2.4 Conclusions

This study is the first to provide longitudinal data regarding the performance of POU filters for Pb removal from private well water. The key finding is that an under-sink AC block filter certified under NSF/ANSI 53 removed influent Pb to very low levels (below the American Academy of Pediatrics' 1 μg/L threshold) during the entire manufacturer stated lifetime (six months) and improved the safety and effectiveness of faucet flushing. Pre-treatment may also be necessary to reach the filter's rated capacity for some wells. The effectiveness of these devices over time has important implications for preventing disproportionate Pb exposure among communities dependent on private well water. Indeed, children relying on private wells have been shown to have a 25% increased odds of elevated blood Pb levels compared to children who receive their drinking water from regulated community water systems (Macdonald Gibson et al., 2020). These areas are often low-income, rural communities and/or minority communities that depend on private wells as a result of historical and ongoing processes of exclusion from municipal services and infrastructure as documented throughout the U.S. (Aiken, 1987; Balazs and Ray, 2014; Colfax, 2009; Heaney et al., 2015; Lichter et al., 2007; MacDonald Gibson et al., 2014; Naman and Gibson, 2015; Seaton and Garibay, 2009; VanDerslice, 2011). Although POU water treatment cannot be considered a turnkey solution to systemic injustices that prevent equitable water access (Vandewalle and Jepson, 2015), this study provides data that can be used to both improve the decision-making of individual well users and to inform evidence-based policies and investments around under-sink POU devices – such as periodic testing events and treatment system subsidies (Zheng and Flanagan, 2017) – to prevent Pb exposures among private well users.

Future research should extend this work to test similar filter designs under wider influent groundwater conditions. Waters with higher hardness, alkalinity, and dissolved organic carbon content may interfere with Pb removal to a greater extent than the waters tested here. Faucet-mounted devices should also be evaluated for private well users. These devices may provide protection from the faucet fixture as a Pb source, but the results of this study with respect to cumulative water usage and Pb loading may not apply to faucet-mounted filters, which have a lower capacity, are challenged by higher first-draw Pb concentrations originating from the faucet, and require additional behavior change to manually bypass the filter when using the hot water. These subtle differences could increase the surface loading and impact the long-term performance of these devices for well users. Finally, AC devices are not appropriate in all scenarios. Other common groundwater contaminants such as nitrate and arsenic are not well removed by AC (USEPA, 2006). Thus, studies of a similar longitudinal nature need to be undertaken for other technologies and contaminants to develop a toolkit of validated solutions for private wells.

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CHAPTER 3: EVALUATING POINT-OF-USE ACTIVATED CARBON FILTERS TO REMOVE PER- AND POLYFLUOROALKYL SUBSTANCES (PFASs) FROM PRIVATE WELL WATER[1]

3.1 Introduction

Per- and polyfluoroalkyl substances (PFASs), including long- and short-chain perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkyl sulfonic acids (PFSAs) as well as replacement perfluoroalkyl ether acids (PFEAs), have generated concern in recent decades for their widespread environmental occurrence and adverse human health effects. Elevated exposure to two legacy PFASs that have been in production since the 1940s, perfluorooctanoic acid (PFOA) and perfluoroctane sulfonic acid (PFOS), has been associated with multiple adverse health outcomes, including liver damage, increased risk of thyroid disease, increased cholesterol, and possible carcinogenic effects (ATSDR, 2018; Grandjean and Clapp, 2015; Steenland et al., 2010). Bioaccumulation of PFASs is also known to occur with longer chain compounds tending to exhibit greater bioaccumulative effects (ATSDR, 2018) and certain PFASs that accumulate in the lungs have been associated with a more severe course of COVID-19 (Grandjean et al., 2020; Pérez et al., 2013).

As a result, long-chain PFCAs and PFSAs 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 limit of 70 ng/L in drinking water for PFOA and PFOS combined (USEPA, 2016a, 2016b). Alternative PFEAs have been developed to replace legacy PFCAs and

[1] This chapter is currently in preparation for submission to the journal *Environmental Health Perspectives*.

PFSAs, however, such as hexafluoropropylene oxide dimer acid (HFPO-DA, also known by its trade name GenX). Research into the toxicity of these compounds to set drinking water regulations in the U.S. is ongoing (Conley et al., 2019; Guelfo et al., 2018; Wang et al., 2017)

Drinking water may contribute a significant portion of overall PFAS exposure, particularly for those relying on private drinking water wells close to 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; 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 PFASs, however, from the cumulative impacts of background sources such as septic systems (Schaider 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 these compounds, interventions are needed to limit drinking water exposures for private well users who may be at risk.

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 and POU reverse osmosis (RO) filters to well owners impacted by the spread of PFASs 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, the National Sanitation Foundation (NSF) released protocol P473 to test and certify household water treatment products for the removal of PFOA and PFOS to below the USEPA health advisory level. Popular outlets such as *Consumer Reports* have also begun to provide independent testing regarding POU filters that may be effective for PFASs (Santanachote and Bergman, 2021) and even make recommendations for maximum limits of PFASs 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 PFASs, 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 for removal of PFOA and PFOS. Performance of pour-through filters comprised of loose granular activated carbon (GAC) 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 for six legacy PFASs associated with aqueous film forming foam (AFFF) from a military base in Colorado. RO units demonstrated effective removal during a seven-day assessment, and small-scale GAC column tests were initially able to remove all PFASs. Lastly, Herkert et al. (2020) provided additional insight into the household performance of POU devices, including ACB filters, in a cross-sectional study of 61 households in North Carolina. RO and dual-stage ACB filters were shown to remove 74–99% of both long- and short-chain compounds in municipal tap waters.

These studies suggest that POU filters may provide an effective solution for private well users concerned about or impacted by PFASs, but important knowledge gaps remain. Anumol et. al. and Patterson et. al. tested POU treatment effectiveness over time but did not evaluate the numerous emerging short-chain perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkyl sulfonic acids (PFSAs) and perfluoroalkyl ether acids (PFEAs) now detected in some drinking waters in the U.S. (Hopkins et al., 2018; Sun et al., 2016). Additionally, these studies were conducted under controlled laboratory conditions, including constant flow rate and water quality, which are not representative of variable household conditions and irregular usage patterns. Small-scale GAC column tests are also unlikely to predict breakthrough of contaminants from ACB filters due to distinct particle size, media conformation, bed volumes, and flow rates. Herkert et. al. tested POU devices under real-world 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 does not provide information regarding the time-to-failure nor account for variations in water usage. Lastly, none of the previous studies included private well water limiting the utility of these studies to provide recommendations to well users because the chemistry of private well water may be very different from the test waters used during NSF certification and can vary widely from one location to another and over time.

To begin to fill these gaps in the scientific and professional community's understanding and to provide the public with improved insight for making risk-management decisions around PFASs in private well water, this study evaluated the performance of 18 under-sink ACB filters installed in the homes of private well owners living near potential PFAS sources for eight months. The key aim was to evaluate the treatment effectiveness of these devices for well users who may be impacted by PFASs from a range of potential sources. By employing a longitudinal study design, performance over time was related to various household-level variables such as volume of water treated, usage patterns, and influent water quality conditions. To date, no other studies have provided a longitudinal analysis of the performance of commercially available POU ACB water filters for PFAS removal under household conditions. These results provide vitally important information toward decision-making in communities impacted by PFASs in drinking water across the nation.

3.2 Methods

3.2.1 Study area and participant recruitment

Eighteen households were recruited to participate from three different communities (A, B, and C) in Orange County and Robeson County, North Carolina, with groundwater quality concerns.[2] Study recruitment and demographic characteristics of each recruitment area have been described in detail previously (Mulhern and MacDonald Gibson, 2020). Briefly, 11 households were recruited in Robeson County approximately five miles west of a current fluorochemical manufacturing facility (cluster C) that had been identified through prior testing by the Robeson County Health Department as impacted by the spread of GenX from the facility. Under a legal consent order, the company was required to provide replacement water supplies to all households with >10 ng/L one of 12 different PFASs, including GenX, in private well water surrounding the facility (North Carolina General Court of Justice, 2019). Households that had previously had their water tested by the local health department but were below this level remained concerned, however, and were identified as potential candidates for this study. No information was known about other PFAS levels among these households. 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. Lastly, four

[2] See Appendix A – Figure A.1.

households were recruited in Orange County near the county landfill, which has been shown to have contaminated nearby drinking water wells but previously had not been tested for PFASs (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 3.1**. Groundwaters in clusters B and C were significantly below the certified pH range. This study was approved by the University of North Carolina Institutional Review Board (IRB Study No. 19-1015).

Table 3.1. Average influent groundwater quality 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
Electrical conductivity (µS/cm)	350	107	168	135	109	35	100–250*
Temperature (°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 CaCO3)	110.5	50.3	30.2	8.8	17.9	7.2	-

*Converted range for 200–500 mg/L total dissolved solids

3.2.2 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 (2,967 L) and time of use (six months). Each replacement cartridge costs $70, representing a maintenance cost of $12/month or approximately $0.02/L if replaced at the recommended intervals. 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 selected filter is a hollow, extruded column produced from a coconut shell carbon mixed with a proprietary ion-exchange metals scavenger and binding agent. Water flows from the outside in with an approximate bed volume (BV) of 0.95 L. 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 that the carbon used has a low overall surface area and high microporosity compared to other commercial activated carbons (**Table B.1**).

Water filters were plumbed in underneath the main kitchen tap 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 usage over time.[3] The data logger's memory could record at 10 sec 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.

3.2.3 Sampling protocol

As described previously (Mulhern and MacDonald Gibson, 2020), samples were collected in 1 L, acid washed HDPE bottles at the influent and effluent sample ports at approximately monthly intervals for eight months (October 2019–June 2020). At the time of each visit, the data logger memory was downloaded and reset. Two sampling months were lost due to Covid-19

[3] See Chapter 2 – Figure 2.1.

restrictions during the study. Since the device did not have a performance indicator built in to alert the user once the volume capacity has been reached, the study was continued two months beyond the recommended six-month lifetime to evaluate the safety of the device in the likely event that cartridges are not replaced exactly at the six month mark. Samples were transported on ice, then stored at room temperature in the dark at the University of North Carolina (UNC) at Chapel Hill until analysis.

3.2.4 Analytical procedures

PFAS analyses were performed using two different analytical methods (I and II) to both verify results and to test for emerging PFASs not included in standard USEPA methods. Appendix B provides a complete list of method analytes (**Table B.3**), detailed analytical procedures, QA/QC protocols, and inter-laboratory comparisons for methods I and II. The two methods are described briefly below.

3.2.4.1 Method I: Solid-phase extraction by USEPA 533

The primary method used for all influent and effluent samples (82 paired samples, 164 total) was anion exchange solid-phase extraction (SPE) and liquid chromatography/tandem mass spectrometry (LC-MS/MS) adapted from USEPA Method 533 (Hunt et al., 2020; Rosenblum and Wendelken, 2019) conducted at UNC Chapel Hill and Research Triangle Institute (RTI) International laboratories. The method includes a standard suite of 24 PFASs (**Table B.3**). 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 ±50% with at least 99% confidence according to USEPA protocols (Munch and Branson, 2004; Rosenblum and Wendelken, 2019). The method detection limit (MDL) was also determined to estimate removals where effluent concentrations were <MRL. The MDL was

defined for each analyte according to USEPA protocols 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.

3.2.4.2 Method II: Large volume direct injection

In addition to the above, nine paired influent/effluent samples (18 total) from the last sampling month from a subset of homes closest to the fluorochemical manufacturing facility in Robeson County (cluster C) were also analyzed for a broader suite of 44 PFASs (**Table B.3**), including several emerging PFEAs known to be associated with the local manufacturer that are not included in USEPA 533 (North Carolina General Court of Justice, 2019), using direct injection of a 200 µL aliquot of sample (i.e., without pre-concentration via SPE) and liquid chromatography triple quadrupole mass spectrometry according to a method developed by North Carolina State University. For this method (referred to here as method II), the MRL was determined as either 1) the lowest calibration standard detected within ±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.

3.2.4.3 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 hours of holding time were determined to be low (<0.05 mg/L) and did not significantly influence the analysis. pH, electrical conductivity, and temperature were measured in the field during each sample event using a handheld probe (HI98129, Hanna Instruments, Smithfield, RI) calibrated daily before use.

3.2.5 Data analysis

3.2.5.1 Statistical tests and mixed effects Tobit regression models

For each household, PFAS removal was assessed by comparing individual and total PFASs in the filter influent and effluent. PFASs not detected above analytical limits in 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 <MRL, several mixed-effects Tobit regression models were fit to the log-transformed concentrations of each individual analyte using maximum likelihood estimation. This was considered superior to a simple substitution approach where analytes <MRL or <MDL are substituted with a value one-half of the limit, due to low levels of many PFASs in influent samples close to the MDL which could result in a low estimation of percent removal and the filter's performance overall for certain analytes, as discussed in a previous assessment of POU filters for PFAS removal (Herkert et al., 2020). Thus, a censored regression model was fit to the data according to the equation:

$$C^*_{i,h} = X_{i,h}\beta^* + \mu_h + \varepsilon_{i,h} \qquad (1)$$

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

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

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 method (Munch and 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 PFASs, PFCAs, PFSAs, and PFEAs) to evaluate: 1) the change in PFAS concentration from the filter influent to the effluent and 2) the effect of well water chemistry (pH, DOC) and cumulative volume of water treated on filter performance. The coefficient β_{filter} in the regressions that identified the change in PFAS concentration between filtered and unfiltered samples was exponentiated to yield an overall percent reduction for each PFAS class such that:

$$\% \text{ removal} = 1 - e^{\beta_{filter}} \tag{3}$$

3.2.5.2 Calculating the sum of PFASs

A common practice during PFAS analysis and risk assessment is to sum the concentrations of all PFAS analytes in each sample. Although this sum can be uncertain due to methods that target only small portion of all possible PFASs, relatively high analytical limits, and the often erroneous assumption that analytes below detection or reporting limits are equal to zero, it is also the basis of emerging regulatory limits for drinking water quality, such as the 20 ng/L threshold recently established for the sum of five ($PFAS_5$) and six ($PFAS_6$) PFASs 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 PFASs (Felton, 2021), and the state of North Carolina implemented a 70 ng/L limit for the sum of 12 PFASs 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 PFASs in each sample to these emerging thresholds, the sum of detectable PFASs was calculated assuming one-half of the MDL (method I) or MRL (method II) for analytes below these limits.

3.2.5.3 Forecasting missing flow data

Due to fieldwork restrictions during the COVID-19 pandemic, sampling visits could not be conducted for two 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 B.1**) was used to estimate the cumulative volume water treated at the end of the study using the "forecast" package in R (Hyndman and Khandakar, 2008).

3.3 Results

3.3.1 Influent PFAS concentrations

Of the 24 PFASs tested for under Method I, eight analytes were detected in the influent above the MRL in at least one study household at least once over the eight-month study duration. Three additional analytes (PFHxA, PFHpA, and PFHpS) were consistently above the MDL in influent samples but never exceeded the MRL (**Table 3.2**). These eleven analytes ($PFAS_{11}$) 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 $PFAS_{11}$ concentrations in influent samples were above the MDL, and 20% were above the MRL. Legacy PFOA and PFOS concentrations were low across all three geographic clusters (max=7.35 and 26.3 ng/L, respectively). PFSAs were detected above the MRL more frequently than PFCAs (37% compared to 3% of the time), but the MRL for the PFSAs was also lower (0.5 ng/L compared to 6.2 ng/L) due to lower levels of background noise from the laboratory environment for these analytes. Relatively high 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. GenX, 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 $PFAS_{11}$ concentrations among all households ranged 4.7–76.4 ng/L (mean=21.4 ng/L; **Table 3.2**). No households exceeded the USEPA Health Advisory Level of 70 ng/L for the sum of PFOA and PFOS, but 34 of 82 samples (41%) exceeded the Vermont and Massachusetts drinking water threshold of 20 ng/L for the sum of five and six PFASs, respectively, and 71% exceeded the *Consumer Reports* recommended threshold for the sum of all PFASs.

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 the short-chain compound perfluoro-2-methoxypropanoic acid (PMPA), which ranged 15.2–99.5 ng/L (mean=53.7 ng/L; **Table 3.1**). The sum of influent $PFAS_{11}$ plus the six additional PFEAs

($PFAS_{17}$) 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 manufacturer, and five of nine exceeded the 70 ng/L limit for the sum of PFEAs (**Figure B.3**) (North Carolina General Court of Justice, 2019).

Table 3.2. Summary of PFAS concentrations above the reporting limit in raw well water representing filter influent.

Type	Analyte	Carbon chain length	Analytical method	*n*	MRL (ng/L)	% >MRL	Mean* (ng/L)	Max inf. C (ng/L)
PFCA	PFBA	4	I	82	6.2	8.5%	33.1	61.7
	PFPeA	5	I	82	6.2	1.2%	6.8	6.8
	PFHxA	6	I	82	6.2	0%	NA	4.1
	PFHpA	7	I	82	6.2	0%	NA	2.9
	PFOA	8	I	82	6.2	4.9%	7.2	7.4
PFSA	PFBS	4	I	82	0.5	79%	3.4	13
	PFPeS	5	I	82	0.5	23%	0.9	1.3
	PFHxS	6	I	82	0.5	72%	1.7	3.9
	PFHpS	7	I	82	0.5	0%	NA	0.4
	PFOS	8	I	82	6.2	8.5%	15.1	26.3
PFEA	GenX	6	I	82	6.2	23%	9.0	14.1
	PFMOAA	3	II	9	5	55.6	7.8	10.8
	PMPA	4	II	9	2	100	53.7	99.5
	PFO2HxA	4	II	9	1	88.9	4.6	10.1
	NVHOS	4	II	9	1	77.8	1.3	1.7
	PEPA	5	II	9	1	100	3.4	6.0
	Nafion BP2	7	II	9	2	88.9	4.6	8.5
	$\sum PFAS_{11}$			82	-	-	21.6†	76.4
	$\sum PFAS_{17}$			9			94.7†	131.3

*Mean of influent concentrations detected >MRL.

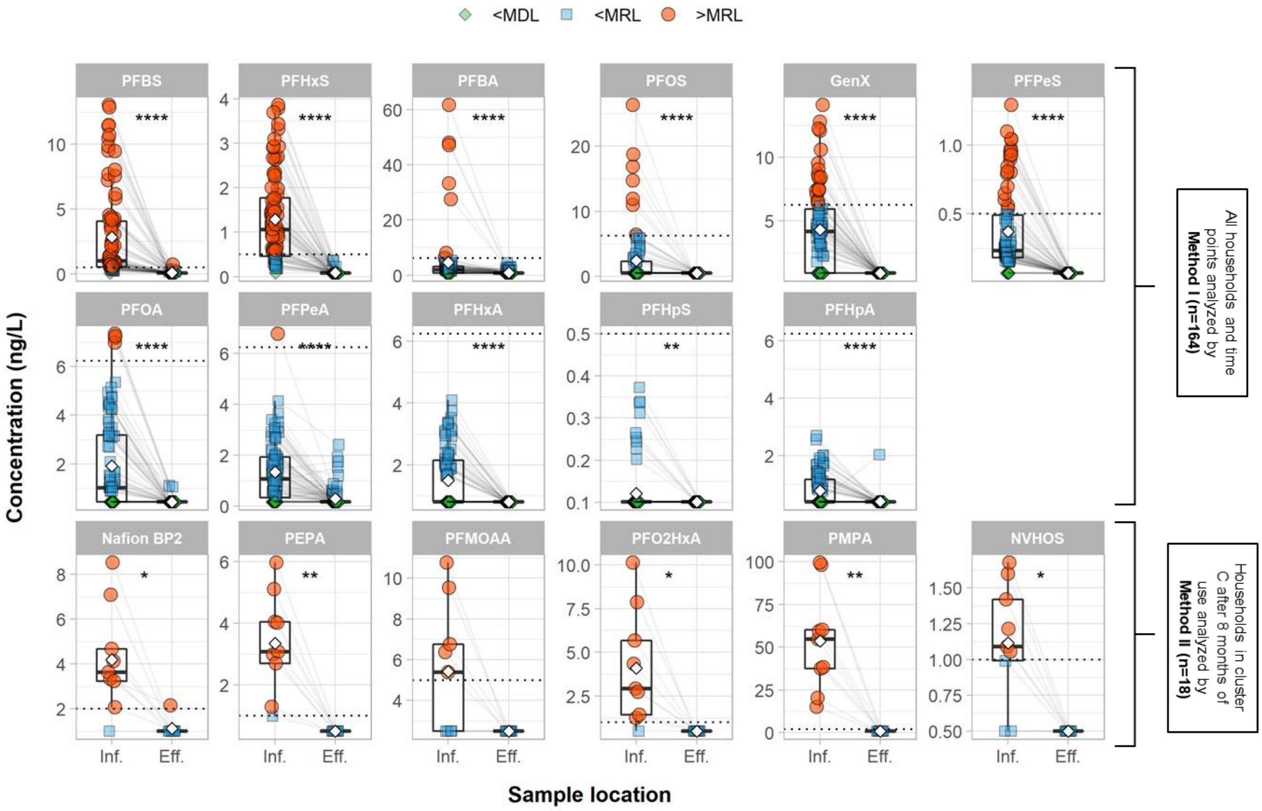

Figure 3.1. Paired filter influent and effluent concentrations of all detectable PFASs demonstrating significant reductions in the effluent concentrations for each analyte. Stars indicate significance of difference between influent and effluent samples from Wilcoxon tests. White diamonds show the censored 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 eight 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.

3.3.2 Reduction of $PFAS_{11}$ in filter effluent

Significant reductions of $PFAS_{11}$ concentrations were observed in the filter effluent for the entire eight-month follow-up (**Figure 3.1**). 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. Only 5.3% of $PFAS_{11}$ analytes in the

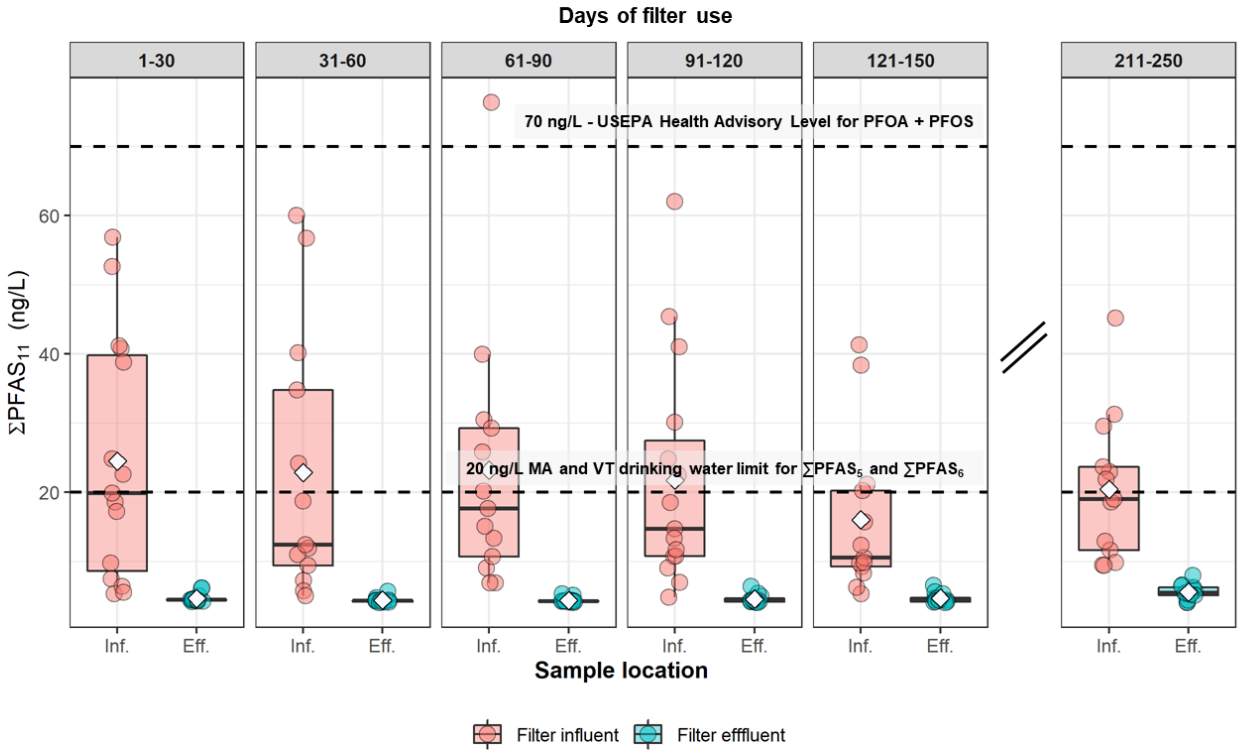

Figure 3.2. Distribution of the sum of $PFAS_{11}$ concentrations in the influent and effluent of all filters at each sample month compared to existing regulatory thresholds for the sum of five and six different PFASs in Massachusetts and Vermont (20 ng/L) and the USEPA Health Advisory Level for PFOA+PFOS (70 ng/L). $\sum PFAS_{11}$ was consistently removed for the entire study period with no significant breakthrough observed.

remaining samples (*n*=164) exceeded the MDL in effluent samples and only 0.1% were above the MRL. Compared to 61% and 20% (respectively) in the influent, this represents a 91% reduction in the prevalence of detectable $PFAS_{11}$ and a 99.5% reduction in reportable $PFAS_{11}$ over the course of the study. The maximum effluent concentration for any single analyte did not exceed 5 ng/L. Only one of 82 effluent samples (1.2%) contained any $PFAS_{11}$ (PFBS) above the MRL, which was detected in the effluent at one household at a concentration <1 ng/L. Using the conservative assumption that all samples <MDL were present at a concentration of MDL/2, the estimated sum of $PFAS_{11}$ in all effluent samples never exceeded 8.2 ng/L and was likely much lower. Paired Wilcoxon signed rank tests on the distributions of influent and effluent samples showed that the effect of the filter on $PFAS_{11}$ concentrations was highly statistically significant,

regardless of chain length (**Figure 3.1**). Consistent removal was also observed at each sampling month, with no significant breakthrough of total $PFAS_{11}$ in the effluent after eight months (up to 250 days) of use (**Figure 3.2**).

3.3.3 Reduction of emerging PFEAs

The nine filters installed in homes in cluster C also effectively reduced 98% of influent PFEAs to below reporting limits after eight months of use (**Figure 3.1**). The compound known as Nafion byproduct 2 was detected in the effluent of one household at a concentration of 2.1 ng/L but otherwise was well removed. 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 reduced 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 in municipal water treatment scenarios (Sun et al., 2016), but were effectively removed by the under-sink ACB filters without contaminant breakthrough even two months beyond the recommended lifetime. All effluent samples were well below the North Carolina legal limits for these PFEAs surrounding the fluorochemical manufacturer of 10 ng/L for each individual compound (North Carolina General Court of Justice, 2019).

3.3.4 Calculated PFAS removal

The mixed-effects Tobit regression models accounting for left-censored data below the MRL showed that the filter effectively reduced all $PFAS_{17}$ concentrations by 97 to 99% (**Table 3.1**). Performance did not vary among the different classes of PFASs, including PFEAs, PFSAs, and PFCAs. The coefficient on the filter term was highly statistically significant ($p<0.00001$) for each model except for the model fit to PFCAs due to the extremely low prevalence of samples with any PFCAs above the MRL (only 1.5%). Regardless, the model estimated removal of >99% for this class. Indeed, 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 reduced all effluent concentrations to below detection, representing 98% removal on average assuming a value of MDL/2 in the effluent (see facet PFBA in **Figure 3.1**).

Table 3.3. Calculated percent removal across the filter for each PFAS type from mixed effects Tobit regression models. *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 >MRL.

Analyte	*n*	% left censored	β	β 95% CI	*p*-value	Est. % removal	% removal 95% CI	% >MRL in effluent
All $PFAS_{17}$	1912	88.1%	-4.22	-5.00 – -3.43	<0.0001	99%	97–99%	0.2%
PFEAs	272	76.5%	-3.50	-4.52 – -2.48	<0.0001	97%	92–99%	0.2%
PFSAs	820	81.6%	-3.91	-4.80 – -3.02	<0.0001	98%	95–99%	0.7%
PFCAs	820	98.5%	-6.65	-	0.95	99%	-	0%

3.3.5 Effect of influent water quality and water usage on $PFAS_{11}$ removal

Mixed-effect Tobit regression models to assess the role of influent water quality and water usage parameters on effluent $PFAS_{11}$ concentrations over time showed no significant effects, indicating a general independence from influent PFAS, influent pH and organic matter levels, competitive sorption effects of DOC, and cumulative volume treated during the recommended lifetime of the filter cartridge (**Table B.2**). Independence from influent pH suggests that removal was dominated by hydrophobic rather than electrostatic interactions, as shown elsewhere (Wang et al., 2019; Zeng et al., 2020). Additionally, over 70% of influent DOC was present in the effluent on average after less than 500 bed volumes BV (**Figure B.4**), but no significant $PFAS_{11}$ breakthrough was observed during the same period, exhibiting an independence similar to what has been shown for other trace-level contaminants through GAC systems (Mulhern et al., 2017).

The number of BV treated was also an insignificant predictor of effluent concentrations. Cumulative water usage through the filter after eight months ranged 210–4655 L (mean=1495 L), or approximately 220–4880 BVs, (**Figures B.1** and **B.2**). Only one household exceeded the manufacturer's recommended capacity. Although no failure was ever detected in terms of contaminant breakthrough, clogging of the filter cartridge reduced the flow rate to unusable conditions in three of 18 households (17%) after only two to three 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 high influent iron or turbidity through the use of a sediment pre-filter (Mulhern and MacDonald Gibson, 2020).

3.4 Discussion

3.4.1 Effectiveness of ACB filters for private well water

This study demonstrates that ACB filters can be an effective option to mitigate legacy and emerging PFAS contamination in private well water. Among the 18 households in this study, under-sink ACB filters effectively removed 97–99% of total $PFAS_{17}$ concentrations for up to eight months. The prevalence of $PFAS_{11}$ above the reporting limit was reduced by 99.5% and the prevalence of reportable emerging PFEAs tested in a subset of households was reduced by 98%. The filter's removal capacity exhibited independence from concerns around short-chain PFASs breaking through earlier and competitive adsorption by natural organic matter during the recommended cartridge lifetime. 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 PFASs 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 PFASs in municipal tap water, where 75–100% of PFSAs, PFCAs, and PFEAs in influent tap waters were removed (Herkert et al., 2020). As was the case here, most compounds were removed to >90%, except for certain analytes where the influent levels were close to the detection limit. Single-stage under-sink filters in the study by Herkert et. al. were not as effective as the filters in this study, with only 29–72% removal of ten PFCAs and PFSAs. This result is unsurprising given the wide range of 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 for PFAS removal under NSF P473. The same is true of refrigerator and pitcher filters, which were also shown to be less effective for PFAS removal (only 29–72% and 36–71% removal, respectively). Faucet-mounted filters showed slightly better removal (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 PFASs in both municipal and private water supplies. NSF P473 certification should be used as a better indicator of effectiveness for PFASs than device design (i.e., under-sink, two-stage, single-stage, etc.).

Additionally, although RO systems are generally considered the safest option for both municipally treated and private well water, ACB filters have several advantages over RO systems for private well owners, including being simpler to maintain, generally have lower capital, operation, and maintenance costs (USEPA, 2007), generate significantly less waste and utilize less water compared to RO (USEPA, 2006), and, when used cartridges are disposed of sustainably, could help prevent PFASs from re-contaminating local groundwater supplies via concentration in septic systems. Three of 18 ACB filters in this study clogged prematurely suggesting that POU or point-of-entry sediment pre-filters may be necessary to extend the capacity of ACB filters for some private well waters. However, RO systems also require significant pre- and post-treatment to protect the membrane and to manage increased corrosivity and negative aesthetic effects of RO-treated water (Patterson et al., 2019; Thomas et al., 2019). RO membranes have also been shown to degrade rapidly (Pratson et al., 2009) and exhibit sporadic breakthrough of up to 25% of influent PFASs during laboratory testing potentially due to leaking membrane seals or poor pre-filter performance (Patterson et al., 2019), suggesting that these devices are vulnerable to failure in household environments. These factors along with the results of this study suggest that under-sink ACB filters may be a more robust, economically sustainable, user-friendly, and environmentally protective household treatment solution for private well users affected by PFASs.

3.4.2 Considerations for application and testing 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 3.1**) 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). 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 practices.

Household testing with higher influent concentrations should also be piloted, although NSF P473 and the average rate of water usage through the filter among households in this study suggest that PFAS loading on the carbon surface is unlikely to exceed the rated capacity of under-sink devices even under much higher influent conditions. For instance, to receive certification for PFOA + PFOS removal under NFSF P473, filters are challenged with a constant influent of 1500 ng/L PFOA + PFOS, added as 1000 ng of PFOS and 500 ng of PFOA, and must maintain a combined effluent concentration of <70 ng/L through 200% of the claimed treatable volume (NSF Joint Committee on Drinking Water Treatment Units, 2016). Thus, in order to claim a capacity of 2967 L during certification, it follows that the filter tested here did not reach 70 ng/L of PFOA + PFOS in the effluent (approximately 5% breakthrough) until at least 5952 L (2 x 2967 L), or approximately 6000 BV. Although NSF P473 does not consider short-chain PFCAs and PFSAs, this rate of breakthrough is comparable to that observed by Zeng et al. (2020) during bench scale experiments with a similar coconut-shell AC treating groundwater with an influent concentration of 156 ng/L for the sum of seven different PFASs. Zeng et. al. showed approximately 45% breakthrough after 25,000 BV, suggesting that 5% breakthrough occurred at approximately 3000 BV.

This comparison suggests that, even with a range of PFASs in the influent, the relevant portion of the theoretical S-shaped breakthrough curve observed during POU treatment is likely to fall within the initial lag period. Variations in time-to-breakthrough for different PFAS types and chain lengths seen during bench-, pilot-, and municipal-scale treatability studies using AC (Rodowa et al., 2020; Sun et al., 2016; Zeng et al., 2020) may not be observed during the relevant treatment window during POU treatment scenarios. On a municipal scale, where the requirement is to treat thousands or millions of gallons per day, the PFAS carbon use rate may only provide one year or less of continuous treatment, representing significant annual costs for a utility (Whitby et al., 2021; Zeng et al., 2020). At the household level, however, where only 1-2 L per person/day is required for direct consumption (USEPA, 2011) and the overall cold water use at the kitchen tap is low (7.6 L/day on average in this study), treatment performance may not necessarily exhibit the same operational dependencies.

3.4.3 Possibilities for PFAS exposure reduction among private well users

The groundwater concentrations in this study were low compared to other communities near PFAS sources (max $PFAS_{17}$=131.2 ng/L). Surrounding fluorochemical waste/manufacturing sites in Minnesota and West Virginia, for example, PFOA concentrations in groundwater have been reported up to 20,000 and 13,300 ng/L, respectively (Hoffman et al., 2010; Xiao et al., 2015). In groundwater near a military base in Colorado, combined PFOA and PFOS concentrations were found up to 1,800 ng/L (Patterson et al., 2019). ACB filters—and POU water treatment in general—may not be appropriate for these worst-case scenarios. Many private well users may be chronically exposed to low concentrations in groundwater, however, where POU treatment could be effectively deployed. As this study shows, even well users close to known PFAS sources such as municipal landfills and fluorochemical manufacturers may still have relatively low levels in well water resulting from complex fate and transport mechanisms in subsurface environments, such as sorption and biotransformation of PFAS precursors, that are not fully understood (Mejia Avendaño and Liu, 2015; Weber et al., 2017). Additionally, groundwater may contain low levels of PFASs even in areas without acute point sources or histories of industrial PFAS use due to diffuse applications of AFFF, leaching from septic systems, and rural applications of biosolids (Lee and Murphy, 2020). Indeed, one effort to document PFAS sources and map drinking water exposure risks from groundwater in Rhode Island suggests that the highest risk regions are likely to be rural areas that may 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 wells above detection, from the impact of onsite wastewater systems alone (Schaider et al., 2014, 2016).

Although some nationwide data are available for PFASs in public water supplies in the U.S. 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 and Murphy, 2020). Of all samples containing detectable PFASs 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 PFASs (Guelfo and Adamson, 2018). A national screening of 163 raw groundwater sources in France also found levels of 1–62 ng/L for 10 different PFASs, comparable to the levels in this study (Boiteux et al., 2012).

In these scenarios, under-sink ACB filters are likely to be highly effective and could significantly reduce chronic exposures and adverse health outcomes among private well users. 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 very low influent PFOA and PFOS concentrations in this study (**Table 3.2**) could still result in blood serum levels up to 0.7 ng/mL for PFOA and 2 ng/mL for PFOS among long term residents from drinking water alone. By comparison, Grandjean and Budtz-Jørgensen (2013) have proposed a serum-based reference dose of 0.1 ng/mL based on reduced vaccine response observed in children, 7–20 times lower than these exposure estimates. Even as research regarding an appropriate reference dose continues for many PFASs (Brown et al., 2020; Guelfo et al., 2018), these data suggest that implementation of ACB filters could reduce such immunotoxic and other health effects associated with chronic, low-level exposures to PFASs from private well water.

3.4.4 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 PFASs 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 PFASs, the fundamental question for many well users—"Is my water safe to drink?"—is fraught with uncertainty. 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, communities 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; Lee and Murphy, 2020; Seltenrich, 2019). The results of this study suggest that under-sink ACB filters certified under NSF P473 could provide a relatively low-cost, widely accessible 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. Individuals may also use these results to make informed decisions about water treatment in their own home, but robust technical and financial support systems are also necessary to effectively implement POU water treatment programs for PFASs and other contaminants among well users (Mulhern et al., 2020). Thus, state and local environmental and public health authorities may also use this information to provide proactive support to communities on private wells that may be at risk of chronic PFAS exposures. Future research should be dedicated to filter assessments in other contexts, as well as for removal of total organofluorine in addition to targeted evaluations of specific analytes (McDonough et al., 2019). Finally, with increasing decentralized treatment for PFASs among both public and private water supplies, life cycle assessments focused on ACB filter cartridges and waste streams may provide important insights into the long-term sustainability and environmental effects of POU treatment programs for PFASs.

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CHAPTER 4: ARE POINT-OF-USE WATER FILTERS SAFE FOR PRIVATE WELLS? EVALUATING THE OCCURRENCE OF MICROBIAL INDICATOR ORGANISMS IN ACTIVATED CARBON BLOCK WATER FILTERS TREATING PRIVATE WELL WATER[1]

4.1 Introduction

Private wells serve the domestic water needs of 42.5 million U.S. (Dieter et al., 2018) and over 4 million Canadian (Statistics Canada, 2017) residents. These wells are vulnerable to a range of chemical and microbial contaminants (DeSimone et al., 2009; Lesage, 2005), but neither the U.S. nor Canada has federal drinking water standards or monitoring and treatment requirements for private well water. Thus, ensuring and maintaining safe drinking water quality is the responsibility of individual well owners. In this situation, private well users may turn to activated carbon point-of-use (AC-POU) water treatment devices as a potential solution. Many consumer AC-POU treatment products advertise the removal of dozens of chemical contaminants, such as lead, volatile organic compounds, and perfluoroalkyl substances, and public information sheets provided by the U.S. Environmental Protection Agency (USEPA) and the National Ground Water Association recommend AC-POU filters as a possible treatment option for well users (NGWA, 2017; USEPA, 2002, 2009). Previous studies have also been dedicated to testing AC-POU devices to remove chemical contaminants from well water (Mulhern and MacDonald Gibson, 2020; Tomlinson et al., 2019).

[1] This chapter is under review in the journal *International Journal of Hygiene and Environmental Health.*

Recommendations from the USEPA and World Health Organization contradict these practices, however, stating that AC-POU filters should not be used with water of poor or unknown microbiological quality (USEPA, 2006; WHO, 2003), which includes many private wells. In a survey of 400 domestic wells across the U.S., 34% were contaminated with total coliforms, a group of bacteria that can indicate potential contamination from human or animal waste (DeSimone et al., 2009). In Virginia, the prevalence of total coliforms was found to be as high as 41% (n=538) (Allevi et al., 2013). Following USEPA recommendations, the manuals of consumer AC-POU devices often make explicit warnings about microbial risks and recommend not using them without disinfection or restricting use to municipally treated water.

These warnings are based on studies—mostly from the 1970s and 1980—reporting inconsistent and sometimes contradictory results, with wide variability in influent and effluent microbial water quality (**Table 4.1**). These studies found that microbial growth within AC filter cartridges and the consequent effects on effluent water quality are difficult to predict and depend on a wide range of factors, including time in operation, influent microbial population, water and ambient temperatures, seasonal trends, stagnation time, rate and frequency of faucet flushing, device design, carbon volume, pre-filter and housing material, and influent nutrient load and organic content. This prior research was also largely conducted on older technologies that used packed granular carbon columns, whereas most modern in-line AC-POU filters use molded or extruded carbon blocks with smaller pore sizes than granular carbon systems and less surface area for microbial growth (CBTech, 2019). Only two of these studies tested AC-POU filters on private well water (Fiore and Babineau, 1977; Snyder et al., 1995), and none assessed the occurrence of coliphages in these systems as a viral indicator of groundwater contamination and health risk (Jofre et al., 2016). As a result, uncertainty remains around whether AC-POU treatment products are safe to use with private wells where the influent water is not guaranteed to be microbiologically safe. This knowledge gap generates considerable confusion around best management practices for private well water and likely deters well users from adopting treatment. Growing recommendations around private well stewardship are focused on encouraging the adoption of testing, treatment, and mitigation behaviors (Flanagan et al., 2018; Malecki et al., 2017), yet in the relative absence of targeted studies to characterize the microbial risks of AC-POU treatment on non-municipally treated water, current knowledge is insufficient to adequately inform health-protective best practice for private well users.

The goal of this research was to compare the occurrence of bacterial and viral indicator organisms in the influent and effluent of AC-POU water filters installed in households on private wells and to evaluate significant water usage and water quality predictors of indicator organism occurrence in the filter effluent. This improved understanding of factors influencing microbial risk provides actionable information for well users, public health practitioners, and policymakers regarding best practices for the safety of POU treatment for private well water.

Table 4.1. Review of selected studies conducted on the microbial effects of activated carbon point-of-use filters.

Study	Water source	Filter type	Study setting	Length of test	# of designs tested	# of filters tested per design	Influent HPC (CFU/mL)	Max effluent HPC (CFU/mL)	Factors affecting filter colonization
Wallis et al. (1974)	Municipally treated tap	Tap-mounted	Lab	6 days	1	1	1	70,000	Time in operation, concentration of assimilable organic carbon within filter
Fiore and Babineau (1977)	Municipally treated tap and 1 private well	Under-sink	Lab and Household	11 weeks	1	6	10 - 300,000	300 - 35,000	Stagnation time, faucet flushing
Taylor et al. (1979)	Municipally treated tap	Under-sink	Lab	24 weeks	4	1	<100	>10,000	Temperature, carbon surface area, flow volume and velocity, time of sampling, influent bacterial population, chlorine removal efficiency of the filter
Smith and Lindsay (1981)	Municipally treated tap	Under-sink and tap-mounted	Lab	55 days	3	2	First-draw: 9,500 Flushed: 160	First draw: 162,000 Flushed: 8,000	Time in operation (flow rate not significant)
Regunathan et al. (1983)	Municipally treated tap	Under-sink	Lab	30 days	1	2	<1 - 330,000	66,000	Stagnation time (no relation between influent and effluent plate counts)
Bell et al. (1984)	Municipally treated tap and untreated groundwater	Various	Lab	5 days	10	Variable, 29 total	10-14,000	350,000	Stagnation time

Geldreich et al. (1985)	Municipally treated tap, dechlorinated	Under-sink	Lab	12 months	4	1	49 - 17,000	84 - 530,000	Time in operation, filter design, time of day, water temperature, competition/inhibition from other bacteria
Reasoner et al. (1987)	Municipally treated tap, dechlorinated	Under-sink	Lab	12 months	7	1	<10,000	260,000	Time of day, faucet flushing, season, temperature, disinfectant residual, unit design, carbon volume, prefilter/cartridge composition, influent bacteria
Snyder et al. (1995)	Private wells and springs	Under-sink	Household	12 months	1	24	<500	First draw: 5,000 Flushed: 300	Influent bacteria, faucet flushing, stagnation time, nutrient load
Chaidez and Gerba (2004)	Municipally treated tap	Under-sink	Household	6 weeks	1	10	10 – 5×10^4	100 – 4×10^7	Organic content, influent water quality, distribution system contamination
Su et al. (2009)	Municipally treated tap	Tap-mounted	Lab	37 days	1	3	20	205	Flow rate, temperature, volume treated per day
Wu et al. (2017)	Municipally treated tap	Tap-mounted	Lab	67 days	1	6	>1,000	>100,000	Presence of chlorinated phenol-based disinfection by-products, presence of pre-filter fabric, operation mode

4.2 Materials and Methods

4.2.1 Study area and recruitment

This study was conducted under real-world conditions in 17 homes with private wells. Participant recruitment for this study has been described previously (Mulhern and MacDonald Gibson, 2020). Briefly, households were recruited from neighborhoods in Orange County and Robeson County, North Carolina, located in three geographic clusters (A, B, and C; **Figure C.1**). Cluster A is a non-agricultural, suburban area 1-2 km southwest from the Orange County landfill. Clusters B and C in Robeson County are peri-urban, mixed-use areas near agricultural activities and confined swine and poultry feeding operations. These areas are also flood-prone and were heavily impacted by hurricanes Matthew in 2016 and Florence in 2018. All wells were within 150 feet of a septic system, and five had surface elevations downgradient of the septic tank (household-specific information available in **Table C.1**). Households were recruited by e-mail, word-of-mouth, and outreach by community partners. This study was approved by the University of North Carolina at Chapel Hill (UNC) Institutional Review Board (IRB Study No. 19-1015).

4.2.2 POU treatment system design

As described previously, an AC-POU water filter was installed below the primary kitchen sink in each household in October–November 2019 (Mulhern and MacDonald Gibson, 2020). The selected filter (A.O. Smith, AO-MF-ADV) is widely available at national hardware stores for $100 and is certified to reduce aesthetic impurities under NSF/ANSI 42 and certain contaminants of health concern, including lead, under NSF/ANSI 53, and two perfluoroalkyl substances under NSF P473. The device is composed of an extruded AC block without a prefilter membrane or fabric and is designed to treat the full flow of cold water at the tap, up to 5.67 L per minute. Sample ports were installed at the filter influent and effluent underneath the sink such that the effluent sample had no interaction with the faucet fixture or aerator (**Figure C.2**). A flow sensor (Sea YF-S201 or Gredia GR-301) and data logger (Onset Hobo State Logger) were integrated into each system to capture water usage patterns.

4.2.3 Monthly sampling

Water samples were collected at the filter influent and effluent monthly from October 2019 to March 2020. Samples were collected in 500 mL sterile HDPE or polypropylene bottles. Before sampling, the sample ports were disinfected with 70% isopropyl alcohol and allowed to dry for a minimum of 30 seconds. Influent and effluent ports were then flushed for 10 seconds prior to sample collection to clear the tubing leading to the sample port and ensure the sample was representative of the true influent and effluent. To ensure proper aseptic procedures, samples were collected at the time of the researcher's visit, meaning that each filter was sampled at a different time of day with varying levels of use and stagnation before sampling. Influent and effluent pH, temperature, and electrical conductivity were measured at the time of sample collection using a handheld probe calibrated daily (Hanna Instruments, HI98219). After sample collection, all bottles were placed on ice and transported to UNC–Chapel Hill within six to eight hours and stored at 4°C until analysis. Most samples were processed within 24 hours, with some samples held for up to 48 hours based on USEPA guidelines (USEPA, 2001).

4.2.4 Water quality analyses

4.2.4.1 Culture-based indicator organisms

Bacterial indicator tests included general indicators of sanitary quality including heterotrophic plate count (HPC) and total coliforms, as well as presumptive *E. coli* as a fecal-specific indicator. Total coliforms and *E. coli* were measured by a USEPA approved enzyme substrate test (Colilert IDEXX, Westbrook, ME) according to Standard Method 9223. Concentrations were recorded as most probable number (MPN) per 100 mL. HPC testing was performed in duplicate via spread-plate using R2A agar according to Standard Method 9215C. Volumes of 0.1 mL were aseptically spread on R2A, and Petri dishes were then covered and incubated at room temperature for 5-7 days. R2A plates were counted manually and reported as CFU/mL. According to the method, high results (>10 colonies/cm^2) were estimated by counting four representative 1 cm^2 squares, taking the average count per square, and multiplying by the plate area. HPC results were analyzed for quantitative variations in plate count and qualitative changes in morphology and color. The number of different colony colors on each plate was quantified as an estimate of the sample richness to characterize changes in alpha-diversity after treatment.

For viruses, F-specific coliphages were selected as the indicator of choice as they can be shed in human feces, are similar in size and morphology to human enteric pathogens, and exhibit similar mechanisms of transport and survival in soils and groundwater (Jofre et al., 2016; Leclerc et al., 2000). Viruses can also be more persistent and migrate further than bacterial pathogens in groundwater and thus may occur in the absence of bacterial indicators (Borchardt et al., 2003; Leclerc et al., 2000; Ogorzaly et al., 2010). F-specific coliphage were enumerated using a single-agar layer assay adapted from USEPA Method 1602 (USEPA, 2001). Briefly, the male-specific coliphage host (*E. coli* Famp, ATCC#700891) was incubated until it reached exponential-phase growth and added to 100 mL of sample pre-mixed with 0.5% magnesium chloride. The sample/host mixture was then combined with 100 mL of 2X tryptic soy agar (TSA) containing ampicillin/streptomycin antibiotic to minimize contamination risks. The sample was mixed and divided into approximately equivalent volumes on five sterile 150x15 mm Petri dishes and incubated at 36.5±0.5°C for 18–24 hours before enumeration. The number of plaques in each plate was summed to give the total number of plaque forming units (PFU) per 100 mL of sample. A method blank using 100 mL of sterile deionized water was included in each batch for quality control. Low levels were detected in the method blanks of some batches (<5 PFU/100 mL) and the blank values were subtracted from the sample result.

4.2.4.2 Bacterial speciation

Dominant colors and morphologies occurring on R2A plates were selected for speciation. Ten colonies were selected that were representative of the dominant colors and morphologies. Briefly, colonies were streaked to isolation on TSA, then inoculated into 1X tryptic soy broth (TSB) and incubated at 36.5±0.5°C. A 500-µL aliquot of the inoculated TSB was mixed with 500 µL of 40% glycerol and sterile water (to achieve a 20% glycerol concentration in the frozen sample), vortexed, and stored at -80±10°C before sequencing. In some cases, colonies did not grow on TSA and were picked from the R2A plates and inoculated into TSB as above. Glycerol stock solutions were sent to a commercial laboratory for DNA sequencing and taxonomical identification (MR DNA, Shallowater, Texas). The Supporting Information (SI) provides details on the sequencing method.

4.2.5 Data analysis

Paired influent and effluent samples for each microbial indicator were analyzed using Wilcoxon signed rank tests. The appropriateness of the Wilcoxon signed rank test was determined by visually inspecting the histogram of the differences between paired sample points for each microbial analyte for approximate symmetry and verified by the Shapiro-Wilk test. To calculate log-removals, households with no detectable indicator organisms for any of the assays were assigned a value of one-half the theoretical detection limit (0.5 MPN/100 mL for total coliforms, 5 CFU/mL for HPC, and 0.5 PFU/100 mL for coliphages).

Multiple logistic regression models were constructed to identify predictors of the odds of each microbial indicator organism occurring in the filter effluent. All models were developed in the software RStudio (R version 4.0.3). **Table C.2** lists the predictor variables evaluated. A searching algorithm was used to select the best models according to the Akaike information criterion (Calcagno, 2020). Significance of predictor variables selected by the algorithm was assessed using Wald tests. Insignificant predictors ($p>0.05$) were removed in a stepwise fashion to reduce model complexity. Predictor variables included in the final model were assessed for multicollinearity using the variance inflation factor and for approximate linearity with the logit of the outcome variable. The random effects of the clustering of data points by household and geographic area were also tested in mixed-effects logistic regression models (Bates et al., 2015). Mixed-effects models were found to result in a zero variance for the household and geographic cluster variables, with negligible effects on the model coefficients, and the structure of the model was reduced to drop the random effects.

4.3 Results and Discussion

4.3.1 Occurrence of microbial indicator organisms in filter effluent samples

4.3.1.1 Total coliforms

Of 66 filter effluent samples collected over the course of the study, five (7.5%) tested positive for total coliforms, representing three of 17 (17.6%) AC-POU filters with a positive total coliform result at any time during the study (**Table 4.2**). No influent or effluent samples tested positive for *E. coli* at any time. The five positive total coliform results in effluent samples ranged 1–2,203 MPN/100 mL. Of these five samples, none of the paired influent samples were positive for total coliform. Six influent samples (9.1%) also tested positive for total coliform (range 1–101 MPN/100 mL) during the study, but none of the paired effluent samples had detectable coliform bacteria.

Table 4.2. Summary of influent and effluent water quality across all 17 households over the course of the study.

Sample location	Analyte	All households *n* households = 17, *n* paired samples = 66			
		mean	***sd***	***range***	***% positive***
Influent	pH	4.93	1.18	3.53–7.35	-
	Temp (°C)	16.7	3.39	9.9–23.4	-
	Electrical Conductivity (µS/cm)	172	128	43–485	-
	HPC (CFU/mL)	1498	4258	<10–25792	82%
	Total Coliforms (MPN/100 mL)	3.40	15.6	<1–101	9.1%
	F+ coliphage (PFU/100 mL)	4.5	6.4	<1–33	55%
Effluent	pH	5.9	1.2	3.6-8.4	-
	Temperature (°C)	18.0	4.3	9.5-28.7	-
	Electrical Conductivity (µS/cm)	166	121	47-461	-
	HPC (CFU/mL)	924	1342	5-9760	97%
	Total Coliforms (MPN/100 mL)	39.6	273	<1-2203	7.5%
	F+ coliphage (PFU/100 mL)	3.5	5.4	<1-30	53%

4.3.1.2 Heterotrophic plate count

Heterotrophic bacteria were nearly ubiquitous in both the filter influent and effluent throughout the study; 82% of all influent samples and 97% of all effluent samples had detectable HPC (**Table 4.2**). HPCs showed wide variability between households and time points. Influent HPCs ranged <10–25,792 CFU/mL (median=108). Mean influent HPC was notably greater in households in cluster A (mean=5,307 CFU/mL) than in B (mean=536 CFU/mL, unpaired Wilcoxon $p<0.0005$) or C (mean=353 CFU/mL, unpaired Wilcoxon $p=0.057$; **Table C.3**). Two households consistently had no detectable influent HPC. Meanwhile, HPC bacteria in the effluent ranged 5–9,760 CFU/mL (median=653). By comparison, effluent HPCs from AC-POU filters treating municipally treated tap water have been recorded 2-3 orders of magnitude greater than these levels (Bell et al., 1984; Chaidez and Gerba, 2004; Geldreich et al., 1985; Wallis et al., 1974; Wu et al., 2017). This may be a function of older filter technologies using granular AC media rather than AC blocks and/or whether the device contains a cloth pre-filter providing additional surfaces for microbial growth. Effluent concentrations were not significantly different between geographic clusters. A statistically significant increase in effluent HPCs was observed in cluster C where influent HPCs were lower ($p<0.001$; **Figure C.3**), but not in clusters A or B.

4.3.1.3 F-specific coliphages

In contrast to the infrequent detections of total coliforms in filter effluent, 35 of 66 effluent samples (53%) tested positive for F-specific coliphage (concentration range 1–30 PFU/100 mL; **Table 4.2**). Prevalence in the filter influent was similar, with 35 of 64 samples (55%) having detectable coliphage. These viruses were detected in 16 of 17 homes (94%) at least once during the study, indicating that nearly all wells were vulnerable to some form of microbial contamination. The small size of virus particles (<100 nm (Lute et al., 2004)) allows them to easily pass through filter pores. When paired samples from all households and time points were aggregated, a statistically significant reduction in the effluent concentrations was detected ($p<0.05$), but the effect size was small (**Figure C.4**). The mean concentration decreased by only 1.13 PFU/100 mL after treatment, and the median influent and effluent concentrations were equivalent (1 PFU/ml). Of the cases with coliphage in the influent, 83% had a lower effluent concentration, with removals ranging from 0.04- to 1.25-$\log_{10}$. However, effluent concentrations increased in 22% of paired samples, representing negative log reductions from -0.22- to -1.45-$\log_{10}$. Thus, a slight attenuation of influent viral coliphage was observed overall, but removal was generally not meaningful to health protection and was highly variable across settings.

4.3.2 Changes in bacterial diversity

The type and diversity of colonies in the effluent were distinct from those in the influent. Influent R2A plates were largely dominated by white, mucoid, and transparent colonies in all three clusters (**Figure C.5**, panel A). White and cream-colored colonies isolated from influent plates were identified as *Ralstonia picketti* and *Bacillus circulans*, respectively. *R. picketti* and *Bacillus* spp. have been implicated in drinking water biofilm formation in diverse environments ranging from industrial and laboratory-based ultrapure water systems to the space shuttle (Adley et al., 2005; Koenig and Pierson, 1997; Kulakov et al., 2002), as well as recognized as opportunistic pathogens associated with nosocomial infections (Alebouyeh et al., 2011; Logan et al., 1985; Ryan et al., 2006). Transparent colonies on influent plates were most likely *Aquabacterium commune*, a bacteria found in biofilms in drinking water utility distribution systems, but not known to be a human pathogen (Kalmbach et al., 1999). **Table C.4** provides DNA sequence BLAST results.

In contrast, yellow colonies dominated effluent plates (**Figure C.5**, panel B). Four yellow colonies from effluent plates were separately isolated. Two of the four were identified as *Sphingomonas paucimobilis,* and the other two were *Cellulomonas xylanilytica,* and *Staphylococcus capitis*. *Sphingomonas* spp. are found in a wide range of aqueous and terrestrial environments with a unique ability to survive in low-nutrient environments and biodegrade organic contaminants (White et al., 1996). *S. paucimobilis* has been identified in biofilms in household settings (such as on shower curtains) and in drinking water in diverse scenarios together with *R. picketti* (Adley et al., 2005; Kelley et al., 2004; Koenig and Pierson, 1997; Kulakov et al., 2002). It has been detected in water supplies in clinical settings and is considered an emerging opportunistic pathogen (Ryan and Adley, 2010). *Staphylococcus* spp. have also been detected in water supplies, including household taps served by private wells (Lamka et al., 1980; Lechevallier and Seidler, 1980), and as human pathogens in clinical settings (Cameron et al., 2015).

Other species, producing pink, red, and orange colonies, also appeared in the effluent plates even when they were not present in paired influent samples. Pink colonies were identified as *Paenibacillus provencensis,* and orange colonies were *Rhodococcus corynebacterioides*, both occurring in a wide range of aqueous and terrestrial environments (Carrasco et al., 2017; Kitamura et al., 2012). Overall, the median number of distinct colors identified on effluent plates increased significantly compared to influent plates (2 in influent vs. 3 in effluent, $p<0.0001$; **Figure C.6)**. This increase in diversity (richness) was observed independent of whether the overall HPC increased or decreased in the effluent (**Figure C.5**).

4.3.3 Factors influencing the occurrence of microbial indicator organisms in filter effluent

4.3.3.1 Total coliforms

Low influent HPC

Low influent HPC appears to have been a factor in allowing total coliform bacteria to proliferate within the filter media in certain households. Four of the five effluent samples that were positive for total coliforms were from filters treating well water with less than the HPC sample median (100 CFU/mL), and all were below the sample mean (1,571 CFU/mL). Evidence from previous research supports the hypothesis that HPC bacteria play a role in preventing the colonization of AC by total coliforms. Camper et al. (1985) demonstrated that when the human enteric pathogens *Yersinia entercolitica*, *Salmonella typhimurium,* and *Escherichia coli* were introduced to virgin granular AC columns in sterile water, all three organisms could form stable biofilms on the AC surface. When the pathogens were introduced to the sterile AC columns together with HPC bacteria, however, the pathogens attached to the carbon surface as before but then rapidly decreased. Similarly, Reasoner et al. (1987) showed that, among AC-POU devices inoculated with bacterial pathogens, including *Klebsiella pneumoniae* and *Aeromonas hydrophila*, the device with the greatest HPC growth demonstrated the most resistance to pathogenic colonization.

The same behavior was observed in household #16 in this study, which had no detectable HPC bacteria in the influent (<10 CFU/mL) and total coliform concentrations as high as 2,203 MPN/100 mL in the effluent after 10 days of use. As a suspected biofilm formed on the carbon surface, shown by the elevated effluent HPCs, the total coliform concentration in the effluent declined exponentially while the effluent HPC remained elevated (**Figure C.7**). Thus, colonization of the filter by native heterotrophic bacteria appears to be protective against the proliferation of coliforms and potential enteric pathogens in the filter media. The multiple logistic regression results confirmed that the influent HPC concentration influenced the risk of total coliforms appearing in the effluent such that each 1-$\log_{10}$ increase in the influent HPC decreased the odds of total coliforms appearing in the effluent by 84% (OR=0.16, 95% CI: 0.01–0.67, $p<0.05$) after controlling for cumulative water use (Model 1, **Table 4.3**).

Low water use

In all three households with coliforms in the effluent, positive samples were only detected in the first few weeks after the filter was installed. In household #16, the total coliform concentration was highest after 10 days (35 L) of use and decreased exponentially (**Figure C.7**), while in the other two households (#15 and #9), concentrations of 6 and 1 MPN/100 mL were detected after 11 and 18 days (34 and 62 L) of use, respectively, and were not detected again thereafter, suggesting that the risk of coliform bacteria may be highest in the absence of a significant autochthonous bacterial community soon after filter start-up. This risk may be exacerbated by low water use. Household #16, for example, demonstrated an extremely low rate of water use from the cold water tap in the first 10 days due to the household's primary reliance on bottled water for most domestic needs (1 L/day after the initial flushing at start-up, compared to the study average of 7.6 L/day), which likely allowed for excessive proliferation of bacteria in the first few days. As a result, the filter in household #16 clogged prematurely, after just 150 L (approximately 40 days) of use (5% of the filter's stated capacity). The maximum flow rate dropped from 3.2 L/min at start-up to 1.3 L/min after 10 days of low use and became unusable after 40 days.

Under laboratory-controlled conditions, Su et al. (2009) showed that low daily use rate and low flow rate both increased the amount of bacterial growth in faucet-mounted AC-POU filters and decreased the filter's lifespan. A use rate as low as 6 L/day reduced the filter's capacity by 26%, and flow rates below 1 L/min increased HPCs in the effluent by up to a factor of 3.5. Coliforms may have been introduced to the filter in household #16 from unsanitary conditions within the household plumbing or during installation and allowed to multiply rapidly due to the lack of use and low influent HPC to prevent their initial growth. Across all households, cumulative water use at the time of sampling influenced the risk of total coliforms in the effluent such that early in the filter's life (less than 50 liters of water treated), the odds of total coliform in the effluent increased by 50 times (OR=51, 95% CI: 3.8–3788, $p<0.05$) after controlling for influent HPC (Model 1, **Table 4.3**). No other water usage or water quality variables, including maximum flow rate, average daily flow rate, influent pH, temperature, or presence of F-specific coliphages, were significant predictors of coliform detection in the effluent.

Particle association

Total coliforms were detected in the influent but not the effluent in six instances, with a maximum observed removal of 99.5% (2.3-$\log_{10}$) after accounting for the Colilert detection limit. Thus, in some cases, AC-POU may significantly reduce influent coliforms. One of the probable mechanisms determining whether coliform bacteria are removed is the extent to which influent coliform bacteria are associated with suspended particulate matter. Although particle association of influent coliforms was not evaluated in this study, as much as 50–100% of fecal coliforms have been shown to be associated with suspended sediment in some groundwaters (Mahler et al., 2000) which may be removed according to the filter's Class I particulate reduction rating under NSF/ANSI 42.

4.3.3.2 Heterotrophic bacteria

Bacterial selection

The filters in this study were selective for species that form suspected biofilms on the carbon surface and outcompete other bacteria for nutrients and attachment sites. Oligotrophic species that are capable of surviving in the low-nutrient environments that may occur during long stagnation periods may persist or increase in the effluent, while other species that are inhibited by competing species may decrease. It is now generally assumed that AC-based devices increase HPCs in the filter effluent (USEPA, 2006). However, the results of this study suggest that it may be more accurate to consider microbial growth within AC-POU filters treating private well water as a shift in the composition of the microbial flora, depending on myriad environmental and design factors, rather than as an increase or decrease in the microbial load.

In this study, the bacterial diversity (richness) of R2A plates increased in the filter effluent (**Figure C.6**), but in other AC-POU filter tests conducted with municipally treated tap water, the overall bacterial richness decreased (Wu et al., 2017). Thus, diversity changes across the filter may also be a function of source water type and the presence of a disinfectant residual. Regardless, the proliferation of heterotrophic bacteria within AC-POU filters is a highly unpredictable process with complex effects on microbial diversity. Depending on the autochthonous bacterial community in the raw well water, other influent water quality parameters, and use patterns, the overall effluent plate count may change significantly in either direction, even after flushing. In cluster C, where influent plate counts were lower, there was a 926% increase in the median effluent concentrations (median influent=58 CFU/mL compared to median effluent=590 CFU/mL; $p<0.001$; **Figure C.3**). Under different groundwater quality conditions in cluster A, median influent HPCs decreased by 11% (median influent=795 CFU/mL compared to median effluent=710 CFU/mL in cluster A; $p=0.095$; **Figure C.3**). After controlling for cumulative water usage, the odds of an increase in HPC in the filter effluent across all households and geographic clusters decreased by 83% with each 1-$\log_{10}$ increase in the influent HPC (OR=0.17, 95% CI: 0.06–0.38, $p<0.001$; Model 2, **Table 4.3**).

4.3.3.3 F-specific coliphages

Influent groundwater quality

Effluent coliphage concentrations were highly correlated with the influent concentrations ($p<0.001$; **Figure C.8**). Although AC has been shown to be capable of virus removal in flow-through column tests (Powell et al., 2000), the optimized conditions necessary for effective removal are difficult to replicate in decentralized water treatment scenarios. Viral adsorption depends on the virus type, carbon surface properties, and water quality parameters, such as pH and ionic strength (Cookson, 1969; Gerba, 1984). Considering that the isoelectric point of F-specific coliphages in water is generally low (e.g., 3.9 for the male-specific bacteriophage MS2

(Dowd et al., 1998) compared to mean influent pH values of 4.3–6.9; **Table 4.2**), most phages in the filter influent in this study were likely negatively charged. The carbon in the filters in this study also likely had a high concentration of negatively charged hydroxyl groups on the surface since a significant increase in the effluent pH was observed (median influent pH of 5.2 to a median effluent of 9.1 at start-up). Thus, a repulsive interaction between like charges on the phage and carbon surface probably prevented significant adsorption from occurring for these waters (Gerba, 1984). Additionally, the influent groundwaters in this study had low ionic strength (6.9×10^{-4}–7.8×10^{-3} M), thus increasing the distance of the electrical double layer around viral particles and increasing these repulsive forces. By comparison, Cookson (1969) showed that optimal virus adsorption kinetics occurred in solutions with ionic strengths of 0.04 – 0.12 M.

Seasonal effects and duration of use

Coliphages were more prevalent in both the filter influent and effluent over time (**Figure C.9**). Thus, the longer each filter was in use, the more likely it was to be challenged by coliphage spikes. One possible explanation is seasonal effects due to lower temperatures, increased rainfall, and infiltration of viruses from nearby septic tanks (Stallard et al., 2021). Another is that viral adsorption is a reversible reaction, thus allowing for possible viral shedding from the filter cartridge after periods of increased occurrence in raw well water (Cookson and North, 1967). This behavior may explain the trend observed in later months where effluent concentrations were slightly higher than influent concentrations (Feb 2020 influent mean=2 PFU/100 mL; effluent mean=5.2 PFU/100 mL) following an increase in the influent concentrations the month before (Jan 2020 influent mean=10.5 PFU/100 mL; **Figure C.9**). As a result of these mechanisms, each week of filter use led to a 18% increase in the odds of coliphage occurring in the effluent when controlling for the influent concentration and cumulative water usage (OR=1.18, 95% CI: 1.05–

1.36, p<0.01). Each 1 PFU/mL increase in the filter influent also increased the odds of a positive coliphage result in the effluent by 28% (OR=1.28, 95% CI: 1.07–1.66, *p*<0.05; Model 3, **Table 4.3**).

Table 4.3. Summary of logistic regression results identifying significant predictors of microbial indicator organisms occurring in the filter effluent.

	OR	95% CI	*p*-value
Model 1: Presence of total coliforms in effluent			
Log_{10} Influent HPC	0.16	0.01 - 0.67	<0.05
Cumulative water usage less than 50 L	51	4 - 3788	<0.05
Model 2: Increase in effluent HPC			
Log10 Influent HPC	0.17	0.06 - 0.38	<0.001
Cumulative water use (L)	1.00	0.99 - 1.00	0.32
Model 3: Presence of coliphage in effluent			
Duration of filter use (weeks)	1.18	1.05 - 1.36	<0.01
Influent coliphage concentration	1.28	1.07 - 1.66	<0.05
Cumulative water use (L	0.99	0.99 - 1.00	0.73

4.3.4 Significance for private well users

The POU water treatment industry has largely developed around controlling chemical contaminants in public water systems, such as lead and disinfection byproducts, but is highly relevant to the needs of private well users. This study provides insight into whether AC-POU water filters may be safely used for private well water treatment in the absence of disinfection. Notably, with one exception, the microbial quality of the effluent of the 17 AC-POU devices tested in this study was not significantly worse than the influent water quality when considering indicator organisms representing gastrointestinal health risk. Indeed, total coliforms were removed from the influent more often than they were detected in the effluent. Effluent HPCs increased in some cases but decreased in others and were similar to or less than the levels in the effluent of AC-POU filter studies conducted on municipally treated tap water. In addition, effluent viral coliphage concentrations were directly related to influent concentrations. Certain bacterial isolates identified as potential opportunistic pathogens were detected in both the influent and effluent, demonstrating that well users are exposed to these bacteria with or without implementing AC-POU treatment. The results of this study thus emphasize the already poor microbial water quality that exists in many private wells, which AC-POU treatment does not significantly improve or exacerbate.

Similarly, Fiore and Babineau (1977) found that AC-POU devices caused both upward and downward fluctuations in HPC and did not affect total coliform levels when tested on five municipal waters and one private well, concluding that AC-POU devices were "microbiologically neutral." Snyder et al. (1995) also showed that total coliforms did not increase in the effluent of any of 24 AC-POU filters installed in homes connected to private wells over one year of use. In fact, the total coliform detection rate in the filter effluent in this study (7.5%) was significantly less than that reported by Chaidez and Gerba (2004) for 10 filters connected to municipal water system, where 82.4% of effluent samples contained total coliforms. These results demonstrate that the potential for colonization of AC-POU filters by total coliforms is not unique to private wells and that choosing to implement household water treatment can alter the microbial quality of both public and private drinking water. The limited studies that exist on private well water suggest that the risk may even be lower for private wells due to the abundance of natural heterotrophic bacteria that may help prevent colonization by unwanted bacteria. These findings suggest that, under normal use conditions, AC-POU filters used to treat private well water do not represent a significant additional risk beyond the existing exposures users may experience from well water alone and that the added health benefits of AC-POU filters to alleviate chemical hazards, such as high lead levels, likely outweigh concerns around microbial changes across the filter.

The results of this study do not indicate an absence of microbial risks associated with AC-POU treatment for private wells, however. In one instance of extremely low water use and low influent HPC, total coliforms colonized the filter, leading to high effluent concentrations (2,203 MPN/100 mL) for a short time. Additionally, coliphages in the influent passed through the filter and potentially shed from the carbon media into the effluent after influent levels subsided, suggesting that these filters provide limited protection from potential viral pathogens. Epidemiological evidence is conclusive that increased ingestion of HPC bacteria in drinking water from AC-POU devices is not a gastrointestinal health risk due to the extremely high infective dose of these species (Allen et al., 2004; Calderon and Wood, 1987; Calderon, 1990; Dufour, 1988; Edberg and Allen, 2004; WHO, 2003). However, certain species, such as *Pseudomonas*, *Klebsiella* and *Aeromonas*, as well as *Sphingomonas* and *Ralstonia* as isolated in this study, can be opportunistic pathogens through alternative exposure pathways like cleaning of wounds, inhalation of water droplets, or cleansing of contacts lenses (Allen et al., 2004; Edberg, 1996; Mena and Gerba, 2009; Rasheduzzaman et al., 2019). Severely immunocompromised individuals could develop greater risk of gastrointestinal illness from heterotrophic bacteria in drinking water, but such conditions are specific and normally require hospitalization (Edberg and Allen, 2004).

Thus, well users who wish to use AC-POU treatment in their home to improve chemical drinking water quality should first take measures to protect the microbial quality of their well. The high F-specific coliphage detection rate (94%) indicates that nearly all wells in this study were vulnerable to some form of fecal contamination and is a significant cause for concern regardless of whether AC-POU treatment is in place or not. Approximately 31% of all waterborne disease outbreaks identified in the U.S. between 1971 and 2006 were associated with consumption of untreated groundwater (Craun et al., 2010). Thus, it is important that state and federal health agencies promote adoption of AC-POU treatment for well users in conjunction with continued efforts to ensure good well stewardship behaviors by caring for "upstream" risks that influence general well water safety and quality. Such behaviors include regularly inspecting the well cap, ensuring adequate separation between the well and all neighboring waste systems, testing annually for total and fecal coliforms and (ideally) coliphages, and shock chlorination of the well if microbial contamination is detected.

4.3.5 Limitations and future study

Recommendations for private well users may be improved through future study of AC-POU devices on a wider range of water quality and use conditions. The results presented here suggest that AC-POU devices do not represent an added risk with respect to bacterial and viral indicators, but follow-up study regarding other microbes such as fungi and protozoa may be informative. Targeted research into the virulence of specific bacterial species identified in the influent and effluent could also inform future decision-making regarding AC-POU devices. Finally, although the microbial water quality did not significantly deteriorate after AC-POU treatment in this study, its use is clearly not adequate to mitigate the existing microbial risks for private well users. Thus, efforts should be made by POU device manufacturers and public health agencies alike to promote a multi-barrier approach to household drinking water treatment among private well users, with an additional disinfection step before or after AC-POU filters to reduce the inherent microbial risks associated with well water.

4.4 Conclusions

Private well users who choose to install AC-POU filters to remove chemical contaminants or improve the water's aesthetic quality should be aware of possible microbial risks and take precautions to minimize them. Recommendations for private well users based on this study include:

- Avoid using AC-POU filtered water for purposes other than drinking, cooking, and washing. For more sensitive needs, such as for large wound irrigation or nasal cleansing (e.g., with a Neti Pot), AC-POU filtered water should be boiled, or an alternative source, such as distilled water or sterile saline, should be used.
- Ensure frequent and consistent use, especially during the first 1-2 weeks after installation and after each successive filter replacement, to allow a healthy biofilm to develop on the filter's surface and prevent any potential coliform bacteria from excessively colonizing the filter.
- Flush the system frequently, especially after the filter has been stagnant overnight or after extended periods of non-use.
- Consider using full-flow, under-sink filters that have higher use rates and flow rates than third-faucet and refrigerator filters providing more frequent flushing and less opportunity for excessive bacterial growth.
- Ensure the microbial safety of private well water through regular testing for total and fecal coliforms and coliphages, well inspections, shock chlorination and/or household ultraviolet disinfection technologies if necessary.

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CHAPTER 5: ASSESSING THE USER EXPERIENCE OF A POINT-OF-USE WATER TREATMENT INTERVENTION FOR PRIVATE WELL OWNERS: IMPLICATIONS FOR EFFECTIVE OUTREACH AND PROMOTING WELL STEWARDSHIP BEHAVIORS[1]

5.1 Introduction

Ensuring the safety of private water supplies is a pressing public health, engineering, and environmental justice issue throughout the U.S. and Canada. Privately-owned wells are estimated to serve the domestic needs of 42.5 million people across the U.S. (Dieter et al., 2018) and 4 million people across Canada (Statistics Canada, 2017). This population can be exposed to elevated concentrations of arsenic (Flanagan et al., 2016a; Walker et al., 2005), lead (Mulhern and MacDonald Gibson, 2020; Pieper et al., 2015, 2018), nitrate (Levallois et al., 1998; Lewandowski et al., 2008), organic chemicals such as pesticides (Gosselin et al., 1997) and per- and polyfluoroalkyl substances (PFASs) (Hoffman et al., 2010; Roostaei et al., 2021), microbial pathogens (Allevi et al., 2013; Stallard et al., 2021) and more (DeSimone et al., 2009). Exposure to both chemical and microbial contaminants from domestic water use may account for a significant burden of disease among certain populations of private well owners (Blackburn et al., 2004). In some areas, racial minorities and low-income communities that are reliant on private wells have also been shown to experience disproportionate drinking water exposures from contaminants such as lead (Macdonald Gibson et al., 2020; Stillo and MacDonald Gibson, 2018), pathogens (Bischoff et al., 2012; Rowles et al., 2020; Stillo and MacDonald Gibson, 2017), arsenic (Rowles et al., 2020), and nitrate (Balazs et al., 2011; Schaider et al., 2019) as a result of

[1] This chapter is under review in the journal *Science of the Total Environment*.

historical and ongoing processes of exclusion from municipal services and infrastructure (Aiken, 1987; Balazs and Ray, 2014; Colfax, 2009; Heaney et al., 2015; Lichter et al., 2007; MacDonald Gibson et al., 2014; Naman and Gibson, 2015; Seaton and Garibay, 2009; VanDerslice, 2011).

Solutions are needed to prevent waterborne exposures among private well-dependent communities. Extending connections to regulated, centralized community water systems may be feasible in some areas but is not economical or practical in many cases (Benavides, 2016). Connection to public water also may not be preferable for some homeowners due to cost considerations (Heaney et al., 2015; Lockhart et al., 2020), lack of trust toward public water utilities (Hu et al., 2011), aesthetic objections (Thomas et al., 2019), or the perception that well water is of higher quality (Fizer et al., 2018; Thomas et al., 2019). Even in areas where municipal service connections may be possible, intermediate solutions are necessary. Bottled water may be a feasible stopgap in some scenarios, but its high cost per liter (up to 1,000 times more expensive than municipal water supplies) may place an unsustainable and inequitable burden on the communities that are most impacted by contaminated well water supplies (Gleick, 2004), and plastic waste from bottled water may have large environmental costs (Hu et al., 2011).

Thus, current research and practice revolves around minimizing the inherent risks and vulnerabilities associated with private well water as the most sustainable water supply for many households. Private wells can still provide high quality drinking water but a holistic "infrastructure for stewardship" is needed to support well owners to maintain drinking water quality and protect health (Fox et al., 2016). Core elements of such an infrastructure include enhanced individual, county, and state-level capacity for well testing, monitoring, and data collection, as well as increased use of decentralized household and point-of-use (POU) treatment interventions. To date, most scholarship on private well stewardship focuses on enhancing well testing to identify the risks that may be present (Colley et al., 2019; Morris et al., 2016; Paul et al., 2015; Renaud et al., 2011; Stillo et al., 2019; Straub and Leahy, 2014). This body of work is one prong of what must be a broader focus on the factors determining each step in the "ladder" of health protective behavior among well owners, particularly on those factors determining decisions to implement household water treatment after water testing has been performed (Fox et al., 2016).

High rates of failure of well testing to translate to water treatment when problems are detected reveal that the determinants of water treatment behaviors among well owners remain opaque. For example, in one survey of private well owners affected by nitrate contamination in Minnesota, 74% of respondents (total *n*=471) reported that they would install some form of water treatment if unsafe nitrate levels were detected in their well, but only 22% of those actually affected by nitrate did so, while 63% either did nothing or switched to bottled water (Lewandowski et al., 2008). Similarly, even in states in the northeastern U.S. where extensive private well testing and outreach programs have been conducted for natural arsenic contamination (Flanagan et al., 2015b, 2016a), many well users still did not adopt treatment. In Maine, 27% of well owners took no action after receiving test results demonstrating arsenic contamination in their wells, while an additional 30% chose to switch to bottled water instead of adopting treatment (Flanagan et al., 2015a). A similar rate of inaction was observed in New Jersey even with required testing under the New Jersey Private Well Testing Act. Among homes where arsenic was measured above the state maximum contaminant level, 36% reported either simply avoiding their well water or taking no mitigation action at all (Flanagan et al., 2018). Additionally, limited data is currently available regarding the experiences of well users with household water treatment devices after they implement treatment, specifically regarding how such treatment may influence long-term decision making around water consumption habits in the home, adherence to continuing treatment, and maintaining water filters. All of these factors determine the overall effectiveness of POU treatment interventions to reduce drinking water exposures (Brown and Clasen, 2012).

As such, the goal of this research was to evaluate the experiences of private well users during a pilot-scale POU water treatment intervention to gain insight into 1) the effectiveness of POU water treatment for reducing well water quality concerns and 2) the perceptions and beliefs that may drive decision-making around adopting and/or continuing POU treatment. Questionnaires and semi-structured interviews were conducted among households involved in a technical assessment of POU treatment devices for well water in North Carolina. The technical results detailing the effectiveness of the devices for a range of chemical and microbial contaminants have been reported previously (Mulhern and MacDonald Gibson, 2020; Mulhern et al., 2021b, 2021a). The results of this work may be used by local and state health agencies as well as nonprofit organizations focused on training and educating well users to inform effective outreach and communication around well stewardship and POU water treatment behaviors.

5.2 Methods

5.2.1 Study area and recruitment

Eighteen well owners were recruited from three separate areas in two North Carolina counties representing distinct socioeconomic and demographic groups (**Table 5.1**). Fourteen households were recruited in Robeson County, a low-income and racially and ethnically diverse area with approximately 17% the population relying on private well water. Robeson County is also ranked the least healthy county in the state of the North Carolina (Population Health Institute, 2019). Study participants were recruited from two areas in the county. Eleven households were recruited approximately five miles southwest of a fluorochemical manufacturing facility that was determined to be responsible for contamination of groundwater with per- and polyfluoroalkyl substances (PFASs) (NCDEQ, 2018; Roostaei et al., 2021). Although the company responsible for the contamination was required by the state to provide replacement water supplies through either bottled water or reverse osmosis water filters (North Carolina General Court of Justice, 2019), the local health department identified some households that would not be considered eligible under the stipulations of the Consent Order and recommended that they be included in this study. These households were recruited by door-to-door visits with employees of the health department. Three additional households were recruited with the help of community partners on the opposite side of the county adjacent to several industrial poultry farms, approximately 15 miles west of the fluorochemical facility.

Four households were also recruited by convenience sampling (e-mail and word-of-mouth) in Orange County from a community in close proximity to the county landfill that has caused concerns around groundwater quality (Heaney et al., 2015). Orange County is predominantly white and middle-class, with approximately 27% of the population relying on well water. Orange County participants were generally higher-income and more highly educated than Robeson County participants (**Table 5.1**). While the sample size was small and cannot be expected to fully elucidate racial and socioeconomic differences, these socioeconomic and demographic differences were important to include as these variables may significantly influence perceptions of POU water treatment and well stewardship behaviors.

Across all three recruitment areas (two in Robeson County and one in Orange County), 21 households were initially invited to collect a water sample to determine whether they may be eligible to participate in the filter assessment. Requirements for eligibility included detectable lead, PFASs, or microbial contaminants at the tap as the three priority contaminants of the technical assessment. After the initial baseline testing, all 21 households were eligible, and 18 elected to receive the filter. Two households had pre-existing whole-house water softeners, but none had previously implemented POU treatment at the tap. Household-specific information is available in **Table D.1**. This study was approved by the University of North Carolina at Chapel Hill Institutional Review Board (IRB Study No. 19-1015).

Table 5.1. Comparison of demographic, socioeconomic, and health indicators among the two North Carolina counties where study participants were located.

		North Carolina*	**Orange County, NC**		**Robeson County, NC**	
			Whole county*	This study (n=4)	Whole county*	This study (n=14)
Income & Poverty	Median household income	$54,602	$71,723	>$50,000†	$34,976	$30,000–$39,000†
	Persons per household	2.52	2.49	2.5	2.81	2.8
	Median home value	$172,500	$308,000	$372,580	$75,600	$69,250
	Poverty rate	13.6%	13.4%	-	31.5%	-
Education	High school or higher	87.8%	92.7%	4 (100%)	77.3%	13 (93%)
	Bachelor's or higher	31.3%	59.7%	4 (100%)	13.7%	0
Race & Ethnicity	White	70.6%	76.9%	4 (100%)	30.6%	6 (43%)
	Black	22.2%	11.8%	0	23.6%	5 (36%)
	Native	1.6%	0.6%	0	42.3%	3 (21%)
	Asian	3.2%	8.1%	0	0.7%	0
	Two+	2.3%	2.6%	0	0.2%	0
	Hispanic	9.8%	8.6%	0	9.2%	0
Health	County health ranking (out of 100)	-	2	-	100	-
	Poor or fair health	18%	14%	-	30%	-
	Low birthweight	9%	7%	-	12%	-
	Uninsured	13%	11%	-	19%	-
Water	% reliance on well water	24%	27%	100%	17%	100%

*Socioeconomic and demographic data from 2019 U.S. Census Estimates (U.S. Census Bureau, 2019a, 2019b). Health data from 2019 U.S. County Health Rankings (Population Health Institute, 2019). Well water reliance data from U.S. Geological Survey (USGS, 2018).
† Median response from categorical household-income questions with five options from <$20,000 to >$50,000 per year.

5.2.2 POU water treatment intervention

This study focused specifically on POU water treatment as a relatively affordable, accessible, and “DIY” solution for well users affected by certain contaminants. Each household was equipped with an under-sink activated carbon block water filter, widely available at national hardware stores and through online retailers, that treats the full flow of cold water at the main kitchen sink (A.O. Smith, AO-MF-ADV). The selected device was certified for removal of lead and PFASs according to protocols set by the National Sanitation Foundation (NSF) and American National Standards Institute (ANSI) (NSF/ANSI 42, NSF/ANSI 53, and NSF P473). The initial retail cost of the filter is $100. It is certified to last for six months, at which point the filter cartridge must be replaced. Each replacement cartridge costs $70, representing an annual maintenance cost of $140 or approximately $12 per month if replaced at the recommended intervals. The performance of these devices was monitored over the course of eight months. The full technical details of the treatment system and water quality testing methods have been previously described in detail (Mulhern and MacDonald Gibson, 2020; Mulhern et al., 2021b, 2021a). The filters were tested two months beyond the recommended manufacturer lifetime due to fieldwork restrictions from the COVID-19 pandemic. At the end of the eight-month evaluation, study participants were given the option to continue using the filter or have it removed from their home. Each household was compensated with a $100 Visa gift card for their involvement in the study. For those that chose to continue using the filter, this compensation was enough to cover the cost of one replacement filter cartridge for another six months, after which the cost of continued maintenance became the owner’s responsibility.

5.2.3 Study participant training and report back

During the study, participants received training from the researchers on how to properly collect water samples from the influent and effluent of the device, how to ensure only filtered water was coming out of the tap for faucets without separate hot and cold controls, and how to change the filter cartridge at the appropriate intervals. Water quality results were reported back to participants through formal letters and graphical reports on three occasions: after the initial baseline test to determine if they were eligible; after the first samples were analyzed two weeks to one month immediately after the installation; and after approximately three months of use. Laboratory shut-downs during the COVID-19 pandemic prevented timely testing and report-back of water samples during the second half of the study. However, as discussed below, the initial early report-back of water quality results made a significant impact on users' perceptions.

5.2.4 Questionnaire design and theory

A questionnaire was used to evaluate the study participants' perceptions about their well water and POU water treatment before and after the study. The questionnaire was first administered with each household on the day that the filter was installed (the "pre-test") and again on the day the study was concluded (the "post-test"). All adult members of each household were invited to complete the questionnaire. The main factors evaluated were perceived vulnerability to drinking water exposures through well water; perceived benefits of POU treatment; perceived self-efficacy in implementing POU treatment, including the ability to acquire reliable information, research available products, select a device, seek help, and install a filter; intent to purchase bottled water, well testing, and POU treatment in the future; and other perceived barriers to POU treatment.

Selection of these factors was informed by Social Cognitive Theory and the Health Belief Model, which focus on how individuals' belief influence action or inaction around a specific health behavior (Bandura, 1998; Rosenstock et al., 1988), and the emerging framework of environmental health literacy, broadly defined as the core knowledge and competencies required to seek out and use information to take actions to reduce environmental exposures (Finn and O'Fallon, 2017; Gray, 2018). These models overlap in their emphasis on self-efficacy as a key component of behavior change (Gray and Lindsey, 2019) and have each been used previously to evaluate health protective behaviors among well users (Colley et al., 2019; Irvin et al., 2019; Straub and Leahy, 2014). Thus, they provide a useful starting point for understanding decisions and behaviors around POU water treatment among well users.

Table 5.2. Factors and questions included in questionnaire delivered to study participants before and after participation in a six-month POU filter intervention. Questions with an asterisk were reverse coded in the scale sums.

Factor	Example Questions
Perceived vulnerability	• I drink my well water when I am at home.* • My well water is safe to drink.* • I feel comfortable drinking my well water.* • My well water comes out of the tap looking dirty.
Perceived benefits	• Treating my well water is important to my health. • Using and maintaining a water filter in my home can protect me from harmful contaminants.
Extended perceived benefits (post-test only)	• Buying water filters to treat my tap water can save me money in the long run. • My tap water tastes better since installing the filter. • I trust my tap water more with the filter installed than before.
Perceived barriers (post-test only)	• Buying replacement water filters to treat my well water is too expensive for me. • Remembering to change out water filter cartridges is too difficult.
Self-efficacy	• If my well water is contaminated, I can do something about it. • I can find reliable information about how to treat my well water. • I can choose the correct type of water filter for my well water. • I can do the plumbing to install a water filter at my kitchen sink.
Intent to purchase	• In the future, I will purchase a replacement water filter for my kitchen sink. • In the future, I will buy bottled water to drink at home. • In the future, I will pay for my well water to be tested to make sure it is safe to drink.

Example questions used to measure each of these factors can be seen in **Table 5.2**; the complete survey is provided in **Table D.2**. All questions were scored using a five-point Likert scale with available responses from 5="Completely Agree" to 1="Completely Disagree" for most questions; 5="Always" to 1="Never" for questions related to the frequency of certain actions (such as "I buy bottled water to drink at home"); and 5="Highly certain" to 1="Not at all certain" for self-efficacy questions. Race, education, and categorical household-income questions from <$20,000 to >$50,000 per year were included (**Table D.2**).

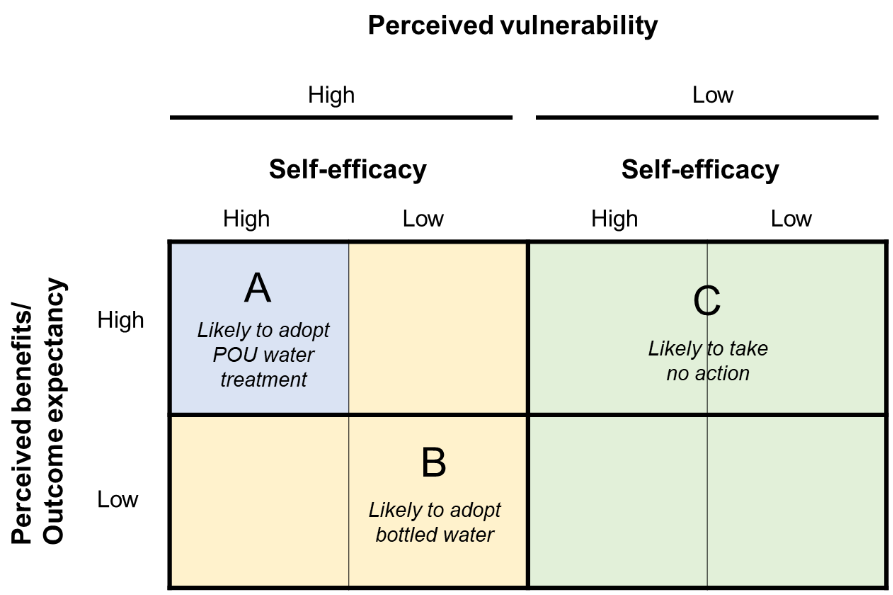

Figure 5.1. Hypothesized theoretical framework used in developing the questionnaire used in this study relying on Social Cognitive Theory and the Health Belief Model.

For the purposes of this exploratory study conducted with a small sample, we relied on a simplified model as a conceptual starting point to understand how these core factors interact to influence well users' decisions and to inform future research questions (**Figure 5.1**). Well users who perceive themselves to be highly vulnerable, have high perceived benefits of POU treatment (also termed "outcome expectancy" in Social Cognitive Theory literature), and have high self-efficacy are theoretically more likely to implement POU water treatment (category A). Well users who perceive themselves to be vulnerable but either do not believe that POU treatment will work or do not believe that they have the ability to implement it will likely adopt an alternative solution such as bottled water, for which the self-efficacy required is negligible and the perceived benefits are high (category B) (Hu et al., 2011; Viscusi et al., 2015). Finally, well users who do not perceive themselves to be vulnerable will likely take no action, regardless of their beliefs around the benefits of POU treatment or self-efficacy (category C). It was hypothesized that most study participants could be classified in category B or C at the beginning of the study as none of them had adopted POU treatment previously.

To evaluate potential cost barriers, we included questions on intent to purchase POU water treatment, bottled water, and water testing in the future using a direct, single-question approach. Since our primary interest was in evaluating participants' internal disposition toward each action rather than real or hypothetical willingness to pay, responses were scored on the same five-point Likert scale from "Completely agree" to "Completely disagree." These questions were each considered separately rather than as items of a single scale as they did not exhibit internal consistency in the pre-test. New questions were also added to the post-test to evaluate other possible perceived barriers (including additional questions around cost, inconvenience, practicality, and perceptions of responsibility over well water quality) and benefits of POU water treatment (such as cost savings, aesthetic improvements, and additional drinking water safety questions) as it became clear that the framework in **Figure 5.1** was unable to account for the study participants' past behavior around POU treatment, as discussed below.

Lastly, questions adapted from the Household Water Insecurity Experiences (HWISE) Scale, a survey instrument for evaluating the breadth of household-level experiences that influence perceptions of water insecurity, were included in both the pre- and post-tests (HWISE

Research Coordination Network, 2019). The HWISE Scale was developed as a cross-culturally appropriate household water insecurity scale validated across 23 countries (Young et al., 2019a, 2019b). The scale includes 12 questions, each representing a different experiential aspect of household water insecurity, including worry, anger, shame, food, handwashing, bathing, washing clothes, sleep, drinking water, interruptions, intermittent supply, and water quantity. Responses on a five-point Likert scale are converted and summed to give a resulting HWISE score from 1-36, with a score of 12 or greater representing a proposed threshold for determining elevated water insecurity (Young et al., 2019a). In this way, water insecurity is defined as the sum of impacts from a range of water-related disruptions. An important limitation of the HWISE Scale is that it was not tested or validated in the U.S. or other high-income countries. To date, however, no other broadly applicable household water security metric is available for high-income countries, making the HWISE Scale an appropriate candidate for adaptation to U.S. contexts.

Table 5.3. Reliability of scales in questionnaires used to evaluate various factors influencing decision-making around POU treatment among well users administered before and after the POU treatment intervention.

	Pre-test (*n*=21)		**Post-test (*n*=14)**	
	# items	Cronbach's alpha (95% CI)	# items	Cronbach's alpha (95% CI)
Perceived vulnerability/ susceptibility	7	0.79 (0.66–0.93)	7	0.88 (0.79–0.97)
Perceived benefits	5	0.71 (0.49–0.93)	4	0.84 (0.71–0.96)
Extended perceived benefits (post-test only)	-	-	12	0.80 (0.65–0.95)
Perceived barriers (post-test only)	-	-	6	0.61 (0.29–0.93)
Self-efficacy	8	0.60 (0.34–0.81)	8	0.79 (0.65–0.94)
HWISE	12	0.82 (0.71–0.93)	12	0.91 (0.84–0.97)

5.2.5 Scale reliability assessment and statistical analysis

In total, 21 individuals completed the pre-test, 13 of whom remained involved for the entire eight-month study period and completed the post-test. One individual also completed the post-test but not the pre-test. The reliability of each questionnaire scale was evaluated using

Cronbach's alpha internal consistency statistic (**Table 5.3**). Cronbach's alpha was calculated for the pre- and post-tests separately since the two samples were not independent. All responses were used in the reliability calculations. All scales had Cronbach's alpha values >0.60 which was used as a minimum threshold for use of the scale in subsequent statistical analyses following established guidance on use of Cronbach's alpha for exploratory research (Hair et al., 2009).

Responses were summed to give a score for each factor (Straub and Leahy, 2014), and the sum of the scores was normalized to the maximum possible score for each scale for comparison between scales. Thus, scores closer to one indicate higher agreement for each factor. Except for the HWISE scale which has a predefined threshold, the scale mid-point of 0.5 was used classify elevated scores for each scale. To assess potential interactions between factors, pre- and post-test results were tested for significant pair-wise correlations using Pearson correlation coefficients. Significant correlations were then subjected to multiple linear regression analysis to control for race, income, gender, and education variables. To analyze changes in participant responses after the study compared to before, the 13 paired responses were analyzed for statistically significant increases or decreases in each scale using paired Student's *t*-tests or Wilcoxon signed rank tests (McDonald, 2014), depending on the normality of the differences in scores between pairs, as confirmed for each scale using the Shapiro-Wilk test. Finally, differences in pre- and post-test scores between participants in different demographic and socioeconomic groups were evaluated using unpaired Student's *t*-tests. All statistical analyses were performed in the software R (version 4.0.3).

5.2.6 <u>Semi-structured interviews</u>

At the study's end, semi-structured follow-up interviews were conducted with seven study participants, representing four households from Orange County, and two households in

Robeson County. Six of the seven interviews were with white participants and one was with a Black participant. These follow-up interviews were an optional component of the study. Open-ended, guiding questions were posed to each participant (**Table 5.4**) to qualitatively understand their experiences using the filter during the study, including their preferences, complaints, perceptions of effectiveness, and potential barriers to long-term adoption of POU treatment. Due to fieldwork restrictions from the COVID-19 pandemic, interviews were conducted and recorded over Zoom. Conversations lasted 30–45 minutes. The recordings were transcribed and analyzed using the software Dedoose to identify recurrent themes.

Table 5.4. Example open-ended questions posed to participants during follow-up interviews at the end of the study.

Example Interview Questions

• Since the filter was installed last November, how often (if ever) did you think about the filter being underneath your sink?
  ○ What were some of those thoughts?
  ○ How did you feel about having it there?
  ○ What came to mind when you thought of it?
• What did/do you like about having the filter installed?
• Was/is there anything you disliked about having the filter installed?
• What do you feel prevented you from buying/using/installing a filter at your sink before the study?
  ○ Why do you think those things prevented you from having a filter?
• Walk me through what you would do if you moved out of state and bought a new house on a well to make sure the water was safe to drink.
  ○ How comfortable would you feel doing these things now?
  ○ How confident would you have been doing these things before?
  ○ How do you feel about your ability to treat your own well water now?

5.3 Results

5.3.1 Factors influencing POU water treatment adoption and user experience

5.3.1.1 Perceived vulnerability

Most study participants perceived themselves to be vulnerable to drinking water exposures through their well water when the study began. In the pre-test, 77% of respondents scored above the vulnerability scale midpoint, and the mean normalized perceived vulnerability score was 0.63. Non-white study participants (n = 8) also had significantly higher perceived vulnerability scores than white participants in the pre-test (mean normalized vulnerability score of 0.81 versus 0.59; unpaired t-test p=0.007). Individual perceived vulnerability scores in the pre-test were moderately correlated with the intent to purchase bottled water (r=0.65, p=0.003; **Figure 5.2**), but not with water treatment. This relationship was significant (p=0.02) even after controlling for race, income, gender, and education suggesting that bottled water is the primary alternative for most well users regardless of race and socioeconomic status.

The mean normalized perceived vulnerability score was significantly lower after the POU treatment intervention (0.63 versus 0.42; paired t-test p=0.0002; **Table 5.5**). Decreases in perceived vulnerability were observed for both low- and high-income participants (**Figure D.1**) and both white and non-white participants (**Figure D.2**). Only 23% of respondents scored above the vulnerability scale midpoint after the intervention (compared to 77% before), indicating a significant reduction in the participants' overall sense of vulnerability to drinking water exposures from well water over the course of the study. One participant described her experience during the recruitment phase of the study in this way:

Table 5.5. Summarized questionnaire results for paired responses before and after the POU intervention. The column "% elevated" indicates the percentage of respondents whose scores were above the scale mid-point or, for the HWISE Scale, the percentage of respondents who were classified as "water insecure" with a score >=12.

	Scale max	Pre (*n*=13)				Post (*n*=13)				Test Sig.
		Mean	Median	Range	% elevated	Mean	Median	Range	% elevated	
Factor scales										**T-test**
Perceived vulnerability	1	0.63	0.71	0.23-0.97	77%	0.42	0.4	0.20-0.89	23%	***
Perceived benefits	1	0.82	0.85	0.60-1.00	100%	0.86	0.90	0.40-1.00	92%	ns
Perceived barriers	-	-	-	-	-	0.44	0.40	0.20-0.70	23%	NA
Self-efficacy – all items	1	0.71	0.70	0.58-0.90	100%	0.79	0.83	0.55-1.00	100%	^
Self-efficacy – research/knowledge acquisition	1	0.72	0.75	0.40-1.00	92%	0.80	0.80	0.60-1.00	100%	ns
Self-efficacy – product selection/maintenance	1	0.70	0.70	0.40-1.00	77%	0.85	0.90	0.60-1.00	100%	**
Self-efficacy – POU implementation	1	0.55	0.60	0.20-1.00	54%	0.65	0.80	0.20-1.00	69%	ns
HWISE	36	8.9	10	2-18	31%	6.4	5	0-21	23%	^
Intent to purchase responses										**Wilcox test**
Treatment	5	3.6	4	3-5	62%	4.2	5	1-5	62%	ns
Bottled water	5	3.0	3	1-5	67%	3.0	3	1-5	60%	ns
Well water test	5	2.9	3	1-4	77%	3.4	3	1-5	85%	ns

Significance levels: ns = not significant; ^ = p<0.1; * = p<0.05; ** = p<0.01; *** = p<0.001

> *When you told me there was something in my water, I had to go and buy cases of water because I was scared for my kids to drink this water and there's something in it…I was cooking [with bottled water] and then I was using it as drinking water. So, a lot of time I would run out of water and I had to go buy more water…Once you came and installed [the filter] I didn't have no problem then, I felt safe.*

Other participants also told the researchers: "*I feel better about using* [*the water*]," "*I'm so glad I don't have to go down to buy water to cook anymore*," and "*If I was cooking and I needed a lot of water like to boil pasta or something I thought, 'hey this water is probably so much cleaner than what I used to use'.*"

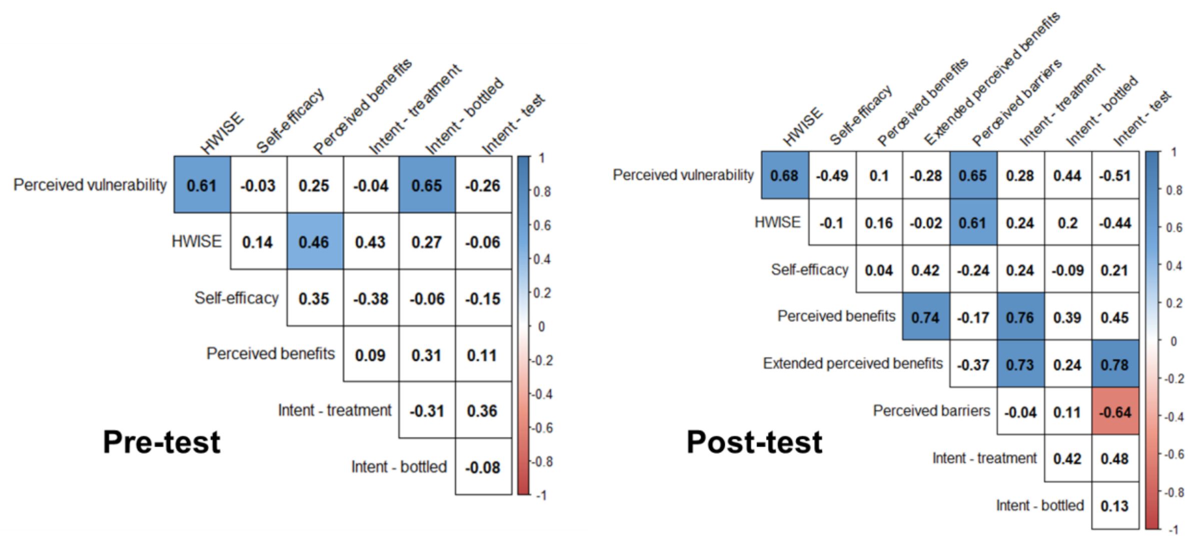

Figure 5.2. Pair-wise Pearson correlation coefficients for factor scores in the pre- and post-tests. Colored cells indicate significant correlations (*p*<0.05).

5.3.1.2 Perceived self-efficacy

Participants scored highly on both the pre-test and post-test regarding their overall sense of self-efficacy (100% of respondents above the midpoint in both assessments; **Table 5.5**). The mean normalized self-efficacy score exhibited only a marginally significant increase after the intervention compared to before (0.71 versus 0.79; paired *t*-test *p*=0.07; **Table 5.5**). Perceived self-efficacy scores were also not significantly correlated with the intent to purchase POU water treatment before or after the study (**Figure 5.2**). Overall, confidence was greatest in the ability to accomplish tasks related to research and knowledge acquisition—such as looking up recommended health limits in drinking water and finding reliable information about problems with well water (92% above the midpoint in the pre-test and 100% above the midpoint in post-test)—and to selecting and maintaining a POU treatment device (77% above the midpoint in the pre-test and 100% above the midpoint in the post test). Participants were less confident in their ability to do the plumbing required to install a POU device (54% and 69% above the midpoint in the pre- and post-tests, respectively).

Shifts in the relative importance of different abilities within the self-efficacy scale were detected after the intervention. One item of the efficacy scale exhibited a statistically significant increase in the post-test: participants became more confident in their ability to "properly maintain a water filter for my home" (mean response of 3.8 versus 4.7; paired Wilcox p=0.012). This question was also highly correlated with the overall score of the efficacy scale and had good item discrimination in the post-test (0.54) but not the pre-test (0.03), suggesting that practical knowledge of how to maintain a POU filter became more important to participants' overall sense self-efficacy over the course of the study. A similar pattern was observed for the question, "I can do the plumbing to install a water filter at my kitchen sink" (item discrimination in post-test of 0.64 compared to 0.06 in pre-test). When asked whether they felt they could do the installation themselves in the future, participants explained, "*Having a sort of hands-on experience with it…I have a much clearer idea of what it would be like to put a filter in place and it's not such a big deal to do*" and "*[The filter] doesn't look like that great big machine I pictured that would be in my crawl space. It's a bit more manageable than in pictured. So I guess yeah, I could do it*."

In contrast, responses to the question, “I can find someone to test my well water to make sure it is safe to drink” exhibited the inverse effect with lower item discrimination in the post-test than the pre-test (0.03 versus 0.41) indicating a shift in participants’ perceptions of the required skills for mitigating well water risks, with a greater emphasis on knowing where and how to obtain information before the intervention compared to practical hands-on knowledge and abilities after the intervention. This shift may also help explain the high self-efficacy scores in the pre-test in that participants may have overstated their confidence in their ability to complete certain tasks before experiencing them.

5.3.1.3 Perceived benefits of POU treatment

Intent to purchase POU water treatment was highly correlated with the perceived benefits after the study, such that participants who perceived the benefits of POU treatment to be high also reported a greater intent to purchase POU water treatment in the future. This correlation was observed for both the condensed benefits scale, which consisted of four questions designed to assess perceptions of health-specific benefits (r= 0.76, p=0.002; **Figure 5.2**), and for the extended benefits scale, which included questions related to cost savings and aesthetic improvement (r= 0.73, p=0.003; **Figure 5.2**). These correlations held true in the post-test results after controlling for race, income, gender, and level of education, but the relationship between perceived benefits and the intent to purchase treatment was not observed in the pre-test. Before experiencing POU treatment for themselves, participants may have answered the questions about the health benefits of POU water filters generically and theoretically, which did not necessarily translate to a personal willingness to buy, install, or maintain a device in their home. After the study, the perceived benefits score was not significantly different than before (0.82 versus 0.86; **Table 5.5**), but it is hypothesized that the perceived benefits became more personally aligned with each individual's goals and motivations when the filter did not clog or cause water pressure issues (see section 5.3.1.4

At the end of the study, 11 of 18 study participants (61%), opted to keep using the filter with the intent to continue to purchase replacement filter cartridges in the future, including three of four participants in Orange County and eight of the 14 participants in Robeson County. Overall, participants indicated a slight increase in the intent to purchase replacement water filters for their kitchen sink after the study, while intent to purchase bottled water stayed the same (**Table 5.5**). This difference was not significant for the group as a whole, but became highly significant when restricted to the 11 households that opted to continue using the filter (mean intent to purchase treatment response of 3.8 before versus 4.7 after; paired t-test p=0.002).

5.3.1.4 Perceived barriers of POU water treatment

The perceived barriers of POU water treatment (measured only in the post-test), such as it being too costly, impractical, ineffective, or inconvenient, were generally low. Only 23% of post-test participants had perceived barriers scores above the scale midpoint, and the mean normalized barriers score was 0.44 (**Table 5.5**). Additionally, the perceived barriers scores were not correlated with the intent to purchase a replacement filter cartridge (**Figure 5.2**), indicating that low intent to purchase treatment was not significantly driven by negative perceptions of it as an intervention overall.

Seven of 18 study participants (39%), one from Orange County and six from Robeson County, chose to have the filter removed from their home at the study's end. The perceived barriers scores were higher among this group than among the 11 participants who chose to continue using their filter (mean normalized score of 0.54 versus 0.39; unpaired t-test p=0.11) while the intent to purchase water treatment was lower (mean Likert score of 3.0 versus 4.7;

unpaired *t*-test p=0.25). These differences were not statistically significant, but it is likely that the sample size was not large enough to reach significance as participants in this group experienced noteworthy barriers not encountered by those who chose to continue using the device. For example, three of the seven had their filters clog prematurely (after only 2-3 months) and likely would require a sediment pre-filter to be able to continue using POU filters effectively. Two households experienced low water pressure with the filter at the kitchen tap even though the filter never clogged. One household had elevated levels of nitrate in their well water, which the activated carbon filter was not designed to remove; this participant was advised to consume bottled water or consider a reverse osmosis unit. Lastly, the seventh household expressed an interest in faucet-mounted POU treatment devices to save room underneath the kitchen sink. Thus, the decision to stop using the filter appeared to be largely associated with technical and practical barriers, which affected participants in both counties and from various socioeconomic groups, rather than with cost barriers.

No significant associations were detected between reported income or education level and the intent to purchase either bottled water or water treatment in either the pre- or post-tests. Nevertheless, income was negatively correlated with perceived barriers after the study, such that lower-income participants reported significantly higher perceived barriers (r=-0.6, p=0.031; **Figure D.3**), and non-white study participants had significantly higher perceived barriers scores than white participants (mean normalized barriers score of 0.58 versus 0.37; unpaired *t*-test p=0.035). Several participants also mentioned cost as a barrier during follow-up interviews when asked why they had never installed a POU filter before. One woman responded:

> *Mainly I thought…buying the bottled water was fine, but also cost. Cost is definitely a factor. I have a very tight budget, so I have to think about every dollar I spend… I always think of the cost and do I really, you know, want to spend a hundred dollars on that?*

This response also reveals the ways in which consumption of bottled water is disconnected from high perceived costs (see van der Linden, 2015). Indeed, this participant reported spending $18–20 per month on bottled water, which is more expensive than the $12 average monthly cost to maintain the filter, but the small, incremental purchases may be more feasible for many households than the upfront costs of POU treatment

Members of one household in Robeson County affected by the spread of PFASs from the nearby chemical manufacturing facility also expressed that it was not their responsibility to install and maintain a water filter in their home because the contamination was the fault of the company: "*They were the ones that induced this stuff in the air and in the groundwater. And so, you know they ought to be responsible for taking care of it. Cleaning it up… If it wasn't caused by them, then it'd be a different story. Then I would feel we should be responsible for our own*."

Lastly, one participant suggested that skepticism of private water treatment companies and conflicting messages was an additional barrier to implementing water treatment:

> *As a homeowner…constantly what you're dealing with is people trying to make money off you. And yeah, you get something for their services, I'm not saying they rip you off, but some people do. And there's a lot of time and effort and research and conflicting opinions…especially if you're not on city water, that's sort of all a part of things*.

Overall, although the perceived barriers of POU treatment reported by study participants were on the lower end of the scale, important obstacles still exist that influence both individual intent and technical feasibility of POU treatment for private wells.

5.3.1.5 Lack of knowledge/awareness of POU treatment

During follow-up interviews, participants frequently expressed that they lacked awareness of POU treatment as an available option before the study. Perceptions that a whole-house system was necessary were common. Several interview participants said that one of the major reasons that they hadn't installed an under-sink filter before taking part in the study was that they did not know under-sink filters were an option or had never thought of it. For example, one interviewee described that, "*First I thought I had to have one on my well, like on the well pump. So that seems overwhelming and expensive. I didn't know that there was a thing I could put underneath the sink like that*." Another explained, "*I hadn't actually thought of [installing a filter]…I just didn't know…what kind of filter we would use…a lot of people I know don't filter their water*." A third household with a defunct whole-house water softener told the interviewer:

> *Honestly, I hadn't thought about an under-sink filter…Originally when I had the whole house [water softener] I thought that would take care of the problem. But there still was a smell and taste to the water even with the whole household, even when it was working properly. So, it would have probably made sense to get a filter at the sink, but I hadn't thought about it.*

Thus, given the high self-efficacy and low perceived barriers scores discussed above, it appears that some well users may not actively decide against POU water treatment as much as simply being unaware that it is a feasible solution to well water quality concerns. This finding also suggests that the self-efficacy scale used in this study, which evaluated skills that were deemed necessary to implementing a POU treatment strategy as determined by the researchers, were not the same skills that participants' themselves perceived that they would need. Indeed, perceived drinking water exposures are unlikely to encourage water treatment behaviors if well users are unaware of the range of treatment options, especially if they perceive some options to be complex, expensive, and difficult to implement, and are unaware of alternative POU solutions that are simple, comparatively low-cost, and for which they already have the necessary skills. This may explain why none of the households had installed a POU treatment system prior to participating in the study, even though 15 of 21 pre-test respondents (71%) indicated a high perceived vulnerability to well water contaminants, high self-efficacy for questions related to

POU treatment, and high perceived benefits/outcome expectancy around POU treatment (category A, **Figure 5.1**). Once they were introduced to POU treatment, however, most study participants chose to continue using the filter and expressed a willingness to continue paying for the intervention.

5.3.1.6 Report-back of study data

Study participants consistently highlighted that the data they received through the report-back letters and conversations with the researcher regarding the filter's effectiveness, not necessarily the presence of the treatment device alone, was what changed their perceptions of their water quality. One participant explained that, although she still preferred the taste of bottled water, "*After getting [the] reports I can see…that it is indeed safe to use for cooking and for drinking…So that's really reassuring and good to know, that it's safe to drink*." Another stated, "*[I'm] glad that [the filter] is doing the job that it's doing. The numbers that we got from you showed that it was working pretty well. So, really glad it's there…there's a sense of relief, you might say…Or in other words, you trust it more*." Even more explicitly, one woman told the interviewer:

> *There was…a mental change about [my water]…You gave me data at the same time [as you tested the filter]…you told me in the beginning about the particles that were in my water, you know, the heebie jeebies. You told me about the viruses, the plastics, the chemicals and the aluminum and what was fine and what wasn't fine and what the filter was treating and what it wasn't so I had that mentality of it being safer. That was the thing that I think primarily got me to go to my water more, was the knowledge that it was safer to drink and it was filtered which was great.*

These quotes emphasize that receiving evidence of the filter's effectiveness was instrumental in increasing the participants' trust in their tap water and decreasing perceived vulnerability to drinking water exposures.

5.3.2 Effect of POU treatment on household water insecurity

The perceived vulnerability scores were positively correlated with the HWISE scores in both the pre-test and the post-test (pre-test r=0.61, p=0.001; post-test r=0.68, p=0.007; **Figure 5.2**), suggesting that the responses to questions in the vulnerability scale around drinking water exposures accurately captured an element of household water insecurity. However, even with the significant decrease in perceived vulnerability (see section 5.3.1.1), some participants expressed still feeling defenseless against well water contamination. One participant said that, even though she was confident in her ability to complete all the tasks on the self-efficacy scale at the study's end, she felt that the problem was still bigger than she could handle:

> *If this is fixing it, if putting a filter on the water that comes out of my faucet right here, if that's the extent of it, great. But what if it's a contamination that affects all the water that comes out of everything that I use? What if I can't take a shower?... I mean what do I do? Do I need a whole house filtration system for all the water that comes out of my well? I don't know if I have the resources to implement something like that. I'm sure there are things that deal with that, that would filter my well water. I just don't know if I can afford that or if I can implement that, you know?*

Similarly, another participant explained:

> *Does [having the filter] mean I have no concerns about my water at all? Not really. Just a week or so ago a neighbor was complaining that suddenly her water seemed…cloudy, dirty or something… and people were speculating 'Could it have something to do with that well being put in just a few houses down?'…I haven't noticed any problems and I'm a little further away. But it just feels like you never know what could change and what else could get into the water that can't be filtered out using the current filter system. You know, stuff that's outside of your control. There's a lot of uncertainty about it and I guess I feel like I don't think [this filter] can really change that very much.*

These experiences highlight the ways in which well ownership itself can be perceived as a threat to household water security, i.e., "the ability to access and benefit from affordable, adequate, reliable, and safe water for wellbeing and a healthy life" (Jepson et al., 2017, p. 3).

This sense of vulnerability clearly extends beyond drinking water exposures at a single tap. The percentage of respondents classified as having elevated water insecurity did decrease slightly from 31% to 23%, but the mean HWISE score was only marginally different after the intervention compared to before (8.9 versus 6.4, paired *t*-test p=0.06; **Table 5.5**). The only item on the adapted HWISE scale used in this study that significantly decreased after the intervention was about how frequently participants worried about their water (mean HWISE item score 3.6 versus 2.5; paired Wilcox test p=0.007). Thus, when paired with water testing and report-back, POU filters may mitigate anxiety related to drinking water safety, but not fully alleviate the lived experience of household water insecurity or the overall liability associated with private well water.

5.3.3 Effect of perceptions of POU water treatment on intent to purchase water testing

Significant correlations were observed in the post-test results between participants' perceived benefits of and barriers to POU water treatment and intent to purchase a well water test in the future. The extended perceived benefits score used in the post-test, including perceived savings, health, and aesthetic benefits, was significantly positively correlated with willingness to get a water test (r=0.78, p=0.001; **Figure 5.2**). In contrast, the perceived barriers score, encompassing barriers of cost, practicality, ineffectiveness, and inconvenience, was negatively correlated with the intent to purchase a water test in the future (r= -0.64, p=0.014; **Figure 5.2**). These associations remained true after controlling for race, income, gender, and education. These results suggest that well owners' perceptions of POU water treatment, including the utility, cost, and health protectiveness, also influence decisions around well testing. Therefore, risk communications designed to promote well testing may also need to confront perceptions around water treatment for private well water to effectively motivate the initial stewardship behavior of obtaining a water test.

5.4 Discussion

This study provides preliminary insights into the effectiveness of POU water treatment for reducing perceived risks and vulnerabilities experienced by private well users and the beliefs that may influence their adoption of POU water treatment to mitigate risks. Notably, a significant reduction in perceived vulnerability to drinking water exposures from well water was observed over the course of the study, especially among low-income study participants, supporting the use of POU filters for reducing perceived well water risks. However, the study also identified key barriers to successfully implementing POU treatment for well users, including lack of awareness of POU treatment options, technical plumbing and water quality challenges, cost, conflicting information, and perceptions of who is responsible for well water contamination. These findings have important implications for non-profit organizations, university researchers, cooperative extension programs, and state and county health agencies developing or investing in programs to serve private well users within their jurisdictions. Most importantly, this study affirms what has been emphasized elsewhere that POU treatment interventions must employ a holistic approach that focuses on the key barriers and facilitators of behavior change among the target population of well users (Morris et al., 2016).

5.4.1 Designing programs to promote POU treatment

In the small sample of well users assessed in this study, a major barrier appears to have been a lack of knowledge or awareness of POU treatment options. Similar knowledge barriers have been reported for private well water testing (Colley et al., 2019; Imgrund et al., 2011; Jones et al., 2005) and other well stewardship behaviors (Kreutzwiser et al., 2011). Some participants perceived that an expensive whole-house water treatment system and/or professional help were required to treat their well water, even though they had a high sense of self-efficacy and perceived high benefits of water treatment before participating in the study. Indeed, professionally installed whole-house water softeners have been shown to be the primary form of water treatment used by private well users in the U.S. and Canada (Jones et al., 2005, 2006; Malecki et al., 2017), indicating that other communities on private well water may experience a similar lack of awareness about POU options. In Texas, households receiving water from a private well were significantly more likely to have water softeners installed than those receiving their water from public water systems, but not more likely to have water filters (Gholson et al., 2018).

This situation often benefits private water treatment companies. Companies may persuade homeowners to purchase a water softener by triggering their perceptions that it will increase the safety of their water (Silvy, 2017), even though water softeners are not certified to remove contaminants of health concern, including metals, organic chemicals, and microbial contaminants (NSF, 2021). In Nova Scotia, private water treatment companies were the leading source of information about well water testing among 420 well owners (Chappells et al., 2015). Dealing with conflicting information from private companies was also mentioned as a barrier by one the participants in this study.

Targeted public education and awareness-raising around POU water treatment as a simple, protective, and affordable option for well users, in contrast to the perceived complexity and expense of whole-house treatment, could thus have a significant impact in dispelling the perception that household water treatment is “out of reach” or requires professional help. Existing messaging around POU treatment for well users may lead to ambiguous notions about household water treatment in general, but often lacks clear, instructional guidance (see NGWA, 2017). Recommendations from trusted sources for specific, validated products (rather than generic information about treatment techniques) to address various contaminants will likely increase awareness and uptake as it eliminates the need to interpret complex information (Frisby et al., 2014) and minimizes the experience of becoming overwhelmed by too many options (Iyengar and Lepper, 2000). Incentive programs, such as free or discounted filters when a well owner tests their well, could provide additional motivation for combined testing/treatment behavior and further reduce the decision-making required of the well owner, although care must be taken to ensure that low-income communities benefit equitably from such programs (Colley et al., 2019; Flanagan et al., 2016b).

Although the factors that motivate or discourage behavior change are highly context-specific and cannot be universally applied (Morris et al., 2016), this study suggests that practitioners ought to consider the possibility that disconnects between theoretically implementing treatment and actually carrying it out as reported around the U.S. and Canada (Flanagan et al., 2015a, 2018; Lewandowski et al., 2008) may be as much a function of lack of knowledge about specific POU treatment options that meet well users' needs as of low self-efficacy, low perceived benefits, or high perceived barriers.

5.4.2 Addressing barriers at each step of POU treatment

Several additional barriers to implementing POU treatment were identified. As past research has shown, environmental messaging alone is unlikely to result in behavior change when "contextual factors limit one's ability to act in consultation with newly acquired knowledge" (Thomas et al., 2019, p. 3131). Thus, four important junctures must be addressed in the design of any effective POU treatment intervention for private well users to enhance their ability to act in response to improved awareness of the need for and availability of POU treatment options: 1) the initial decision to adopt/purchase a POU device; 2) the installation of the device/physical alteration of the household environment; 3) the recurrent decisions to use the filtered water for domestic uses, including drinking and cooking, over other sources such as bottled water or other un-filtered taps in the house; and 4) maintaining the filter properly by changing the cartridges at the appropriate intervals. **Figure 5.3** shows each of these steps and summarizes our suggestions for programming and future research.

First, the success of this study was ultimately a function of a close relationship of trust developed between the study participants and the researchers from a well-respected public university in the state, including monthly contact for almost one year from the initial recruitment to the end of the study. As Morris et al. (2016) have pointed out, a credible and trusted messenger is a critical component of effective outreach to private well owners. This relationship allowed study participants to ask questions, voice their concerns, and receive hands-on training and technical support. As a result, practical "how-to" knowledge of POU treatment, such as doing the plumbing and changing the cartridge, became more important to the participants' overall self-efficacy after the study. Participants may have overstated their ability to perform actions necessary to implement POU treatment on the self-efficacy scale in the pre-test (Koc, 2021; Kruger and Dunning, 1999) reinforcing that knowing *how* to perform well stewardship behaviors, not only *what* behaviors are necessary, is an important determinant of action (see Kreutzwiser et al., 2011).

Thus, programs that provide personalized support from trusted sources are much more likely to succeed. Indeed, the problems experienced by participants in this study that led seven of the 18 to stop using the filter were all related to technical challenges rather than cost concerns.

Examples of technical support programs that could be adopted for private well water management in the U.S. include water and wastewater treatment circuit riders and external support programs that provide technical, financial, and administrative assistance around a specific jurisdiction. Such programs have improved functionality and water quality of rural and other small water systems, and similar programs could be designed to support private well users (Miller et al., 2019). Currently, very few states have policies or programs around well water testing or maintenance and none have any mandate for water treatment (Bowen et al., 2019). Similarly, local health departments vary widely in terms of the services they provide to well users and their capacity to respond to well water concerns (Wait et al., 2020). State and federal funding to equip local health departments to deliver POU treatment and maintenance services and trainings in addition to testing could have a significant impact. Efforts must be made to prioritize low-income areas with free and/or subsidized services that may perceive higher barriers to implementing treatment on their own, as shown in this study.

The results of this study also make clear that the potential of POU technology to alleviate perceived vulnerabilities and increase users' trust of their tap water depended largely on pairing the filter with testing and report-back of water quality results. Collecting and sharing data on the filter's technical performance was thus a powerful reinforcing mechanism for the targeted behavior of prioritizing the filtered water. Such individualized testing and report-back may not be possible for large-scale POU treatment interventions, but efforts should be made to provide clear recommendations around the use of POU treatment that increase confidence in the treated water quality based on water quality testing during field assessments (see Flanagan et al., 2015a; Mulhern and MacDonald Gibson, 2020; Mulhern et al., 2021a; Powers et al., 2019). To date, such evidence-based validations of POU treatment for private well water are lacking (Malecki et al., 2017) but are critical toward disseminating information that positively reinforces treatment behaviors. Lastly, motivations for replacing the filter cartridge at the appropriate intervals to maintain high quality water in the filter effluent must considered. This study was not designed to evaluate this last step of the behavior chain, but appropriate incentives could be developed. One possibility is to establish take-back programs, either by manufacturers or local governments, that provide coupons or economic rewards for each filter change and would also generate environmental co-benefits (Atasu et al., 2009).

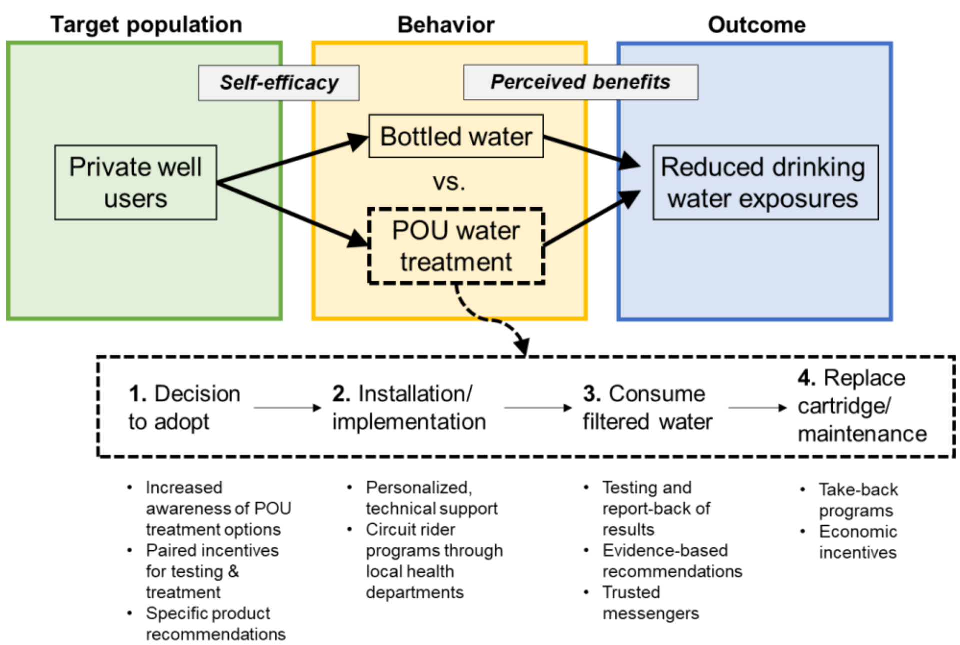

Figure 5.3. A simplified behavior model adapted from Rosenstock et al. (1988) identifying the key steps required to implement POU water treatment for well users compared to bottled water.

5.4.3 Implications for well water testing

Implementing a cohesive POU water treatment intervention in this manner at a local or regional scale may also benefit well water testing efforts. Importantly, this study identified a significant relationship between well users' perceptions of the barriers and benefits of POU water treatment with their willingness to obtain a well water test in the future. Well owners may not want to know the results of a water test if the barriers to a treatment solution are perceived to be prohibitively high, for example. Conversely, they may be inclined to test if they perceive the barriers of implementing treatment to be low and believe that treatment could effectively solve any potential problems revealed. Previous research has shown that people often only change their behaviors around tap water consumption if they believe they have a viable alternative, indicating that presenting the risks alone without information about available options may not be effective (de Franca Doria et al., 2005). Additionally, several previous studies have indicated that the perceived costs of well water treatment may have a negative effect on testing behavior among well users in North Carolina (Stillo et al., 2019), Wisconsin (Malecki et al., 2017), the Northeast (Straub and Leahy, 2014), and Ontario, Canada (Jones et al., 2005), but previous empirical assessments of risk communications to increase well testing in the U.S. and Canada have not addressed perceptions of POU treatment (Paul et al., 2015; Renaud et al., 2011).

This study suggests that such communications and outreach for well owners may be more effective when conducted holistically, addressing testing and treatment behaviors together. Well owners who perceive themselves to have a greater sense of control over problems with their well water have also been shown to be more likely to test and perform well maintenance (Schuitema et al., 2020). As Bandura notes, "Successful efficacy builders…structure situations for people in ways that bring success and avoid placing them in situations prematurely where they are likely to fail" (Bandura, 1998). Thus, promoting well testing alone without support for interpretation of the results and implementation of treatment may not be an effective mechanism of developing sustainable well stewardship behaviors, especially in low-income communities.

5.4.4 Strengthening systems for private well stewardship

Despite the potential for POU water treatment to significantly reduce real and perceived drinking water exposures from well water, the participants in the POU intervention tested in this research made clear that POU treatment is not a cure-all solution for the risks and liabilities experienced by private well users. Indeed, POU treatment may not be protective against microbial contaminants in well water (Mulhern et al., 2021b) and/or fail to mitigate exposures through other routes such as dermal contact and inhalation during showering, which may be significant from some contaminants like volatile organic compounds (Brown et al., 1984). Water treatment at a single household tap or even multiples taps also cannot alleviate concerns around water quantity from wells running dry or pump failure (cf. Lockhart et al., 2020). Critiques of the equitability of POU water treatment as a technological fix to water insecurity among low-income and environmental justice communities in the U.S. are thus warranted (see Vandewalle and Jepson, 2015). If not paired with equitable, well-resourced, and long-term support programs, emphasis on decentralized POU treatment may merely exacerbate preexisting inequalities around drinking water quality. Indeed, it is the very absence of legal and political structures to support private well users that creates and preserves experiences of water insecurity among marginalized populations in the U.S. and Canada (Meehan et al., 2020). Thus, improving state and local capacity to deliver broad public health services to private well users is essential (Sabogal and Hubbard, 2015). Positive examples include the U.S. Centers for Disease Control and Prevention's Safe WATCH program, which aims to support local public health departments to strengthen programs for private drinking water systems (CDC, 2019), and legislation in North Carolina designating emergency funds for capital-intensive treatment and remediation for well owners (NCDEQ, 2017).

5.4.5 Study limitations

This study had several limitations. First, these findings are based on a small sample of well users in North Carolina and thus only provide preliminary insight to be used as the basis for future research and program evaluation. The most effective practices surrounding water treatment promotion must account for place-specific characteristics such as socioeconomic factors, the history of contamination, and institutional trust. The participants in this study were recruited from areas already aware of local groundwater contamination issues and thus may have markedly different perceptions, beliefs, and barriers than other communities. Additionally, although the results presented here provide valuable insight to user experience, this study was primarily designed to evaluate the technical treatment effectiveness of POU devices rather than the drivers of behavior change. Active researcher invovlement with the study participants may have had unknown influences, including but not limited to social desirability bias in questionnaire and interview responses. The questionnaire data was also limited by the fact that the theoretical framework used to design the pre-test did not address perceived barriers beyond self-efficacy. Thus, the perceived barriers scale was only administered in the post-test and could not be compared to baseline perceptions before the study.

Nonetheless, this study provides important insights related to mitigating risks of exposure to contaminants among North American populations relying on private wells. Importantly, no empirical studies regarding the effectiveness of health promotion and risk communication on POU treatment adoption and compliance with maintenance recommendations among private well users are currently available. Conducting large-scale intervention studies among North American private well users represents an important future research need. As part of this future work, formal investigations of willingness-to-pay for POU treatment among well users would also be informative for policy development.

5.5 Conclusions

This study sheds light on the beliefs of private well users that influence POU water treatment behaviors and the effectiveness of POU water treatment to reduce perceived drinking water exposures from well water. After the eight-month POU water treatment intervention, participants reported feeling significantly less vulnerable to drinking water exposures in their well water. Before the study, participants perceived that well water treatment required professional help and a whole-house system, suggesting that lack of knowledge of POU treatment as an available and effective solution was a significant barrier. After the study, most participants decided to continue using the POU treatment device and generally reported positive perceptions of POU water treatment, including high perceived benefits and low perceived barriers. Perceptions of POU water treatment after the study were also significantly correlated with intent to purchase a well water test in the future, suggesting risk communication around well water testing should also address perceptions of treatment. POU water treatment cannot be considered a solution to adverse drinking water exposures nor household water insecurity in isolation, however, and must be paired with long-term public health services and support programs for private well users to be effective.

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CHAPTER 6: CONCLUSION

6.1 Summary of Findings

This study provides important insights into the effectiveness of POU water filters for private well users. As discussed in the Introduction, multiple barriers, or “off-ramps,” to continued stewardship behaviors (such as POU treatment) exist for private well users after receiving the results of a water test (**Figure 1.1**). These barriers include a lack of evidence regarding the effectiveness of POU devices for certain priority chemical contaminants in private well water, such as lead and PFASs; uncertainty surrounding the microbial safety of POU devices for well water; and limited insight into the perceptions that drive decision making around adoption of POU treatment among well users.

The preceding chapters address each of these major barriers with respect to activated carbon block (ACB) POU filters, a common technology in consumer water treatment products. Importantly, this research is the first to provide data on the longitudinal effectiveness of ACB POU filters for lead and PFASs in well water. As demonstrated in Chapter 2, filters removed 98% of all influent lead on average and significantly improved the safety and effectiveness faucet flushing for lead mitigation at the tap. Filters consistently reduced lead levels at the tap to below the American Academy of Pediatrics recommendation of 1 μg/L of lead in drinking water, thus providing valuable evidence in favor of their use to prevent childhood lead exposures from well water.

Similarly, as discussed in Chapter 3, the filters removed 99% of all influent PFASs for up to two months beyond the manufacturer recommended lifetime of the device. This finding is highly relevant to the many communities around the U.S. and Canada that are impacted by PFAS contamination of groundwater from various potential environmental sources. Given that PFASs are still a relatively new drinking water contaminant, considerable uncertainty existed around whether consumer POU treatment products would be effective for emerging compounds for which no treatment certifications have been developed. The results of this work increase confidence in the utility of ACB POU devices to mitigate PFAS exposures and may be used to inform proactive policies and remediation plans. Specifically, in an ongoing contestation surrounding the fluorochemical manufacturer in southeast North Carolina, this information may be used by the North Carolina Department of Environmental Quality and Robeson County Department of Public Health to inform the types of water treatment devices recommended and made available to impacted homeowners in the area (North Carolina General Court of Justice, 2019).

When making public recommendations around POU water treatment for well users, however, state and local agencies must also take care to promote and provide technical assistance for additional well maintenance and stewardship behaviors to protect the microbial quality of well water (Simpson, 2004). As shown in Chapter 4, microbial indicator organisms, which point to the possible risk of pathogens and vulnerability of private wells to leaks, seepage, and possible influence from nearby septic systems and other hazards, were detected in 94% of the study participants' wells. It was determined that the presence of microbial indicator organisms in untreated well water does not necessarily indicate an increased microbial risk for ACB filters. Under normal conditions of use, filters neither improved nor exacerbated the microbial water quality. However, well users must also take preventive action to minimize existing microbiological risks. POU devices may be recommended to treat for chemical contaminants in well water but should be done so in conjunction with holistic well stewardship education to address chemical and microbial risks in parallel.

In support of broader use and promotion of POU treatment among well users, Chapter 5 detected a significant decrease in well users' perceived vulnerability to well water contamination after using the filter for up to eight months, but a general lack of knowledge around POU water treatment interventions prior to the study. This finding suggests that POU filters can be effective to improve the lived experience of private well users, but that one of the possible causes of well water tests failing to lead to treatment behaviors is that well users are unaware of the options available to them. One of the reasons for this lack of knowledge may be the lack of research in this area, affirming the importance of further applied research to inform best practices for well users. Additionally, participant responses to questionnaires revealed that perceptions of the benefits of and barriers to POU treatment were significantly correlated with intent to obtain a well water test in the future, indicating that the steps included in the cycle of well stewardship behaviors (**Figure 1.1**) are highly interconnected. Thus, it was hypothesized that improved adoption of well stewardship behaviors, including testing, treatment, and maintenance, may occur when messaging around risks and solutions are paired.

6.2 Policy and Action Recommendations

Given estimates that 300,000 new private water wells are drilled each year, private water supplies promise to remain a public health concern in the future (US General Accounting Office, 1997). The combined impact of this work may benefit communities across the nation attempting to address the multifaceted and overlapping challenges of private water supplies, corrosion of pipes and lead leaching, PFAS contamination, and microbial safety of groundwater supplies. Multiple stakeholders can take specific actions in response.

6.2.1 Individual well users

Under-sink ACB POU filters certified under NSF/ANSI 53 and NSF P473 may be purchased by individual well users as a relatively affordable option for reducing lead and PFASs in well water. Well testing is recommended to verify the levels of influent contaminants to ensure that they are not outside the concentration ranges under which they were tested and validated. Additionally, ACB filters are not appropriate for certain well water contaminants such as nitrate and microbial pathogens. Thus, installing an ACB filter should not supersede the necessity of obtaining a well water test and performing additional well management behaviors such as wellhead inspections and maintenance.

At the same time, testing may be difficult to access in rural communities, is prohibitively expensive for emerging contaminants such as PFASs, and can be highly inconvenient, whereas ACB POU treatment products can easily be obtained at a local hardware store or online and installed at the kitchen sink without professional assistance. Thus, well users who have reason to be concerned about particular contaminants in their area, such as PFASs, due to proximity to likely sources or other testing performed in the area, may implement treatment as a proactive, preventive measure and obtain testing for certain contaminants as it becomes available or affordable.

6.2.2 Local Health Departments

In North Carolina, General Statute 87-97 requires local health departments to have private well programs that permit, inspect, and provide testing for new wells (NC General Assembly, 2008). Although the range of services that local health departments provide to well users beyond the minimum permitting and inspection requirements varies widely, such as the availability of reduced price testing options, all counties across the state already provide some form of private well services. In a review of the current state of these services and disparities across the state, Wait et al. (2020) showed that 97% of health departments already provide some technical assistance for well users when contacted, and 83% regularly provide on-site assistance at private residences. Most health departments (76%) received daily inquiries from well users in their county.

Based on this research, it is recommended that local health departments across North Carolina integrate evidence-based recommendations related to POU treatment for lead and PFASs into their existing communications with well users. Updated websites with recommendations for treatment products certified under NSF/ANSI 53 (for lead) and NSF P473 (for PFASs), including the specific device tested in this study (AO-MF-ADV, A.O. Smith), could significantly impact uptake of POU interventions and reduce exposures. With additional state and federal funding through programs such as the U.S. Centers for Disease Control and Prevention's Safe WATCH program (Sabogal and Hubbard, 2015), local health departments with a high proportion of well users in their jurisdiction could strengthen and expand their well water programs to train local staff members to provide onsite technical assistance for POU treatment installation and maintenance, as well as offer subsidized treatment products to incentivize broader well testing (see Chapter 5).

6.2.3 State agencies

The North Carolina statute mandates that new wells be tested for arsenic, barium, cadmium, chromium, copper, fluoride, lead, iron, magnesium, manganese, mercury, nitrates, nitrites, selenium, silver, sodium, zinc, pH, and bacterial indicators, but not PFASs (NC General Assembly, 2008). Based on the results of this study and others (Chapter 3), it is clear that private wells are vulnerable to the spread of PFASs in the environment from myriad potential sources and that private well users may be chronically exposed to these contaminants at low levels. Thus, PFASs should be included in the testing mandate for new wells to strengthen the infrastructure for testing for these contaminants, and POU treatment should be recommended as a proactive measure to reduce potential PFAS exposures among this population.

6.2.4 Physicians

In the U.S., children whose parents are on Medicaid are required to have blood lead tests performed at 12 and 24 months (107th Congress, 2001). Such screening efforts by health care providers generally lead to recommendations for well water testing, but clear indicators of blood lead poisoning among children, ought to be paired with immediate recommendations for interventions. This work provides a robust evidence-base for the effectiveness of ACB filters certified under NSF/ANSI 53 for reducing water lead levels at the tap among private wells, and can be confidently recommended by pediatricians who recognize clear signs of blood lead exposure among children on well water. As researchers in the field of pediatrics have pointed out, "The news media and Internet are the sources of information that may be anxiety provoking to parents. The pediatrician has the responsibility to be knowledgeable about some of these concerns and should be able to provide answers if there are adequate data that allow a definitive conclusion" (Brent and Weitzman, 2004, p. 1172). This principle applies to private well use as a risk factor for young children and POU treatment as an effective intervention, akin to recommendations around wearing a helmet when biking or gun safety in the home (Brent and Weitzman, 2004). Well testing should also be recommended but can be done so in parallel with effective, easily implementable treatment recommendations. In fact, such an approach may be more effective to encourage testing behavior (Chapter 5).

As recommended elsewhere, general practitioners serving populations with a high prevalence of private well use should also inquire about drinking water sources and potential environmental exposures when documenting a patient's medical history (Charrois, 2010). Although there are cases where well users should consult with qualified well water technicians or water treatment professionals, in some cases, such as during screening tests for blood lead poisoning, this work and future studies like it could serve to equip physicians to make informed recommendations as a trusted source, potentially leading to enhanced uptake of POU treatment among well users, rather than directing users to private water treatment companies.

6.2.5 Researchers

This work provides policy-relevant conclusions through intentionally evaluating POU water treatment amidst real-world complexity where multiple problems coalesce. Thus, this study offers a model for other researchers focused on private well water risks and exposures to develop research that follows a solution-oriented (rather than problem-oriented) paradigm.[1] A litmus test for solution-oriented research developed by physicians focused on reducing childhood obesity can also be applied to the issues surrounding private well water exposures (see Robinson and Sirard, 2005). Following guidance given to medical students and residents regarding screening and diagnostic tests for delivering patient care, these researchers suggest that a study only be performed if:

1. It can be known what will be concluded from each possible result, and,
2. The result could directly change the steps taken to address a clinical, policy, or public health problem.

[1] See Chapter 1: Introduction – Section 1.3.2

This study met these criteria in that, like a clinical diagnostic test, it led to precise and actionable recommendations to improve the health of well users that may also be used to implement policy changes. These types of prospective, experimental studies are often costly compared to retrospective or observational designs, but if environmental engineers, exposure scientists, epidemiologists, and others who seek to improve drinking water quality among the almost 50 million people who rely on private wells in the U.S. and Canada follow these guidelines, research efforts may quickly converge around protective solutions.

As part of this effort, a continued focus on the structural mechanisms driving disparities in access to drinking water in the U.S. and Canada is important to inform and design interventions to test. As others have pointed out, a "rich understanding of how disparities in access to safe drinking water are produced and maintained is essential for understanding environmental justice concerns and developing effective public health interventions" (Balazs and Ray, 2014, p. 603).

6.3 Ongoing Research

This study included additional research goals not included in this dissertation that are ongoing. First, in addition to testing the under-sink ACB filter, a field test of POU ultraviolet (UV) light emitting diodes (LEDs) for in-line disinfection as a polishing step after filtration was conducted. This application of UV LED technology is the first of its kind and further field verification of these devices for private water supplies could contribute to technological improvements and broad implementation with potential reductions in gastrointestinal illness among private well users. Additionally, an analysis of the cost-effectiveness of the ACB water filters for long-term use based on the estimated health benefits accrued from reduced lead exposures is also ongoing. This work will provide supplementary decision-making information for the promotion and adoption of POU devices to reduce childhood lead exposure among well users.

6.4 Limitations & Future Study

There are several limitations of this work that have been discussed in the preceding chapters. First, even though this longitudinal study is the first of its kind among well users, the sample size of 18 well users was small. Not only did this sample not encompass the full range of possible influent groundwater conditions which may influence treatment effectiveness, it also limited the study design to testing only one treatment device. Large-scale prospective trials of POU treatment interventions for lead, PFASs, and additional chemical and microbial contaminants would further justify the use of POU treatment as an effective intervention for well water at scale and inform the development of targeted state and federal policies. The inclusion of various POU treatment product designs and technologies—including POU ultraviolet disinfection in addition to under-sink ion-exchange, membrane, ceramic, and activated carbon filters—is essential to developing a robust inventory of validated devices for private well users who experience various risks. This study focused on lead and PFASs and previous work has been dedicated to arsenic (Zheng, 2017), but few solutions for nitrate and microbial pathogens have been tested among private wells. Validation of treatment solutions for these contaminants in private well water thus remains a research priority.

Additionally, although the study participants' perceptions and beliefs around POU insights provided valuable insights, the findings were limited by the observational nature of the study and the researcher's active involvement with the participants, potentially introducing a social desirability bias to the results. Thus, these results are preliminary in nature and ought to be tested empirically through risk communication trials to assess adoption of treatment when promoted alone, promoted in parallel with testing and/or when paired with treatment subsidies. Future studies such as these that bring a solutions-oriented research paradigm to bear on the issues of private well water and health will continue to provide important, actionable information to policymakers, public health professionals, and public agencies charged with serving private well users.

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APPENDIX A: SUPPLEMENTARY MATERIAL FOR CHAPTER 2

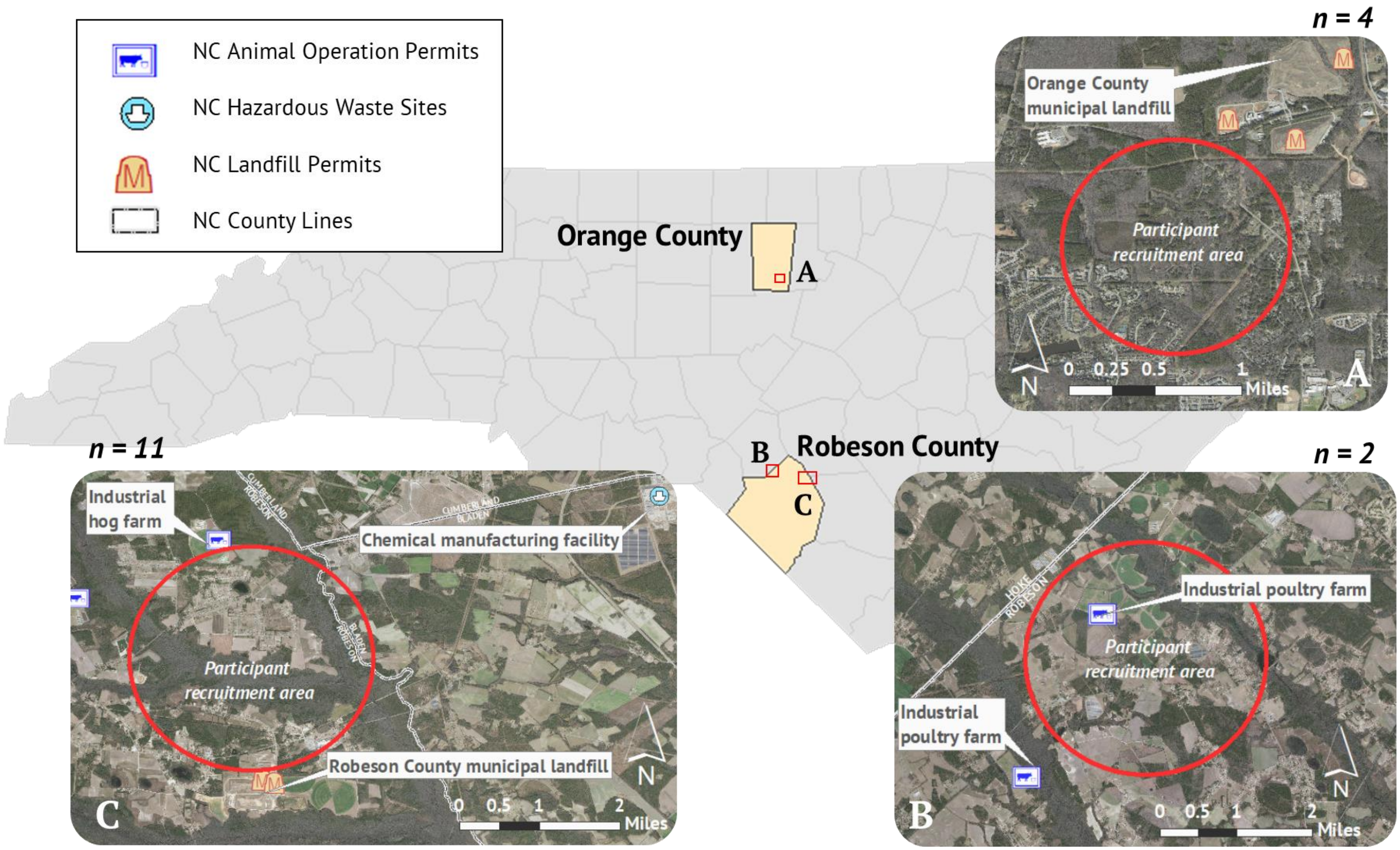

Figure A.1. Map of study participant recruitment areas across three geographic clusters (A, B, and C) in Orange County and Robeson County, North Carolina

Table A.1 Household-specific information of study participants within each geographic cluster.

House ID	House type	Year of home construction	Year of well construction	Well depth (ft)	Baseline Pb – 250 mL first draw from faucet (µg/L)	Preexisting treatment
Cluster A						
5	Single family home	1985	Unknown	Unknown	2.72	Water softener
8	Single family home	1985	Unknown	Unknown	1.57	Water softener
9	Single family home	1987	1987	145	0.13	None
10	Single family home	1990s	Unknown	Unknown	0.41	None
Cluster B						
19	Single family home	1972	Unknown	25-30	34.33	None
21	Single family home	1955	1955	26	0.40	None
Cluster C						
1	Manufactured home/trailer	2010	Pre-2000	35	9.93	None
2	Single family home	2000	2000	Unknown	26.26	None
3	Manufactured home/trailer	1993	1996	35 ft	7.99	None
4	Manufactured home/trailer	1990s	2000	35	14.00	None
7*	Manufactured home/trailer	1976	Pre-1995	Unknown	0.69	None
11	Manufactured home/trailer	2002	Unknown	Unknown	2.83	None
13	Manufactured home/trailer	2018	2000	25-30	5.47	None
14	Single family home	1998	2014	75	8.46	None
15	Single family home	1997	Unknown	Unknown	20.27	None
16*	Manufactured home/trailer	1970s	Pre-1995	Unknown	-	None
17	Single family home	1983	1986	Unknown	9.36	None

*Two homes connected to the same well

Table A.2. Influent groundwater quality of each participating household compared to the required water quality conditions for POU Pb removal certification according to NSF/ANSI 53. pH, electrical conductivity, temperature, DOC, and alkalinity measurements are averaged from monthly influent samples from each household. [Ca2+], [Mg2+], [Cl-], and [SO42-] show single baseline measurement values.

House ID	pH	Electrical conductivity (μS/cm)	Temp. (°C)	DOC (mg/L)	Ca2+ (mg/L)	Mg2+ (mg/L)	Hardness (mg/L CaCO3)	Cl^- (mg/L)	$SO4^{2-}$ (mg/L)	CSMR	$HCO3^-$ (mg/L)	$CO3^{2-}$ (mg/L)	Carbonate Alkalinity (mg/L CaCO3)	Langelier Saturation Index
NSF/ANSI 53	6.5 ± 0.25 8.5	<180	20 ± 2.5 20	>1.0	-	-	10-30 100	-	-	-	-	-	10-30 100	
Cluster A														
5	6.73	212	17.2	0.75	29.9	1.35	80.2	5.45	7.64	0.71	75.48	0.0	61.9	-1.6
8	7.78	457	17.1	1.46	15.3	5.30	60.0	17.9	2.66	6.73	196.38	0.6	162.1	-0.5
9	7.09	452	17.7	1.64	70.5	3.88	192.1	14.8	9.20	1.61	197.44	0.1	162.2	-0.5
10	6.75	281	17.0	0.78	35.9	4.90	109.8	14.2	8.59	1.66	82.43	0.0	67.6	-1.5
Mean	**7.09**	**350.24**	**17.23**	**1.16**	**37.89**	**3.86**	**110.54**	**13.10**	**7.02**	**2.68**	**137.93**	**0.20**	**113.47**	**-1.05**
SD	0.43	107.01	0.25	0.40	20.25	1.54	50.32	4.63	2.58	2.37	59.03	0.25	48.71	0.54
Cluster B														
19	4.29	359	18.0	0.44	10.4	3.20	39.1	36.7	1.08	34.04	0.12	0.0	0.1	-7.3
21	6.56	76	18.5	1.88	5.54	1.85	21.4	4.49	3.00	1.50	17.70	0.0	14.5	-3.1
Mean	**5.43**	**217.30**	**18.25**	**1.16**	**7.96**	**2.52**	**30.25**	**20.59**	**2.04**	**17.77**	**8.91**	**0.00**	**7.31**	**-5.23**
SD	1.13	141.55	0.23	0.72	2.42	0.68	8.84	16.10	0.96	16.27	8.79	0.00	7.21	2.12
Cluster C														
1	4.90	76	17.6	0.34	2.73	0.627	9.4	3.42	1.53	2.23	0.44	0.0	0.4	-6.7
2	4.48	169	19.1	0.79	8.40	2.15	29.8	12.4	9.23	1.34	0.18	0.0	0.1	-7.0
3	4.23	115	18.4	0.28	5.62	3.81	29.7	8.60	1.05	8.19	0.04	0.0	0.0	-8.1
4	4.41	124	21.5	0.22	4.10	3.11	23.0	6.60	1.17	5.62	0.05	0.0	0.0	-7.9
7	4.26	134	18.4	3.40	2.48	1.21	11.1	11.8	28.5	0.41	0.13	0.0	0.1	-7.9
11	4.30	118	18.5	0.37	5.95	2.38	24.6	13.7	1.05	13.09	0.09	0.0	0.1	-7.7
13	4.70	54	18.6	0.36	3.66	1.46	15.1	6.87	1.24	5.52	0.16	0.0	0.1	-7.2
14	4.25	89	18.3	0.27	3.45	1.90	16.4	9.50	1.62	5.85	0.07	0.0	0.1	-8.0
15	4.55	54	20.0	0.32	2.92	1.68	14.2	6.98	1.21	5.77	0.43	0.0	0.4	-7.0

16	4.71	130	17.7	0.29	2.48	1.21	11.1	11.8	28.5	0.41	0.19	0.0	0.2	-7.3
17	3.93	131	17.6	0.32	2.61	1.47	12.6	9.48	12.7	0.75	0.03	0.0	0.0	-8.9
Mean	**4.43**	**108.66**	**18.68**	**0.63**	**4.04**	**1.91**	**17.92**	**9.20**	**7.99**	**4.47**	**0.16**	**0.00**	**0.13**	**-7.62**
SD	0.26	34.51	1.12	0.89	1.80	0.87	7.19	2.94	10.37	3.76	0.14	0.00	0.11	0.61

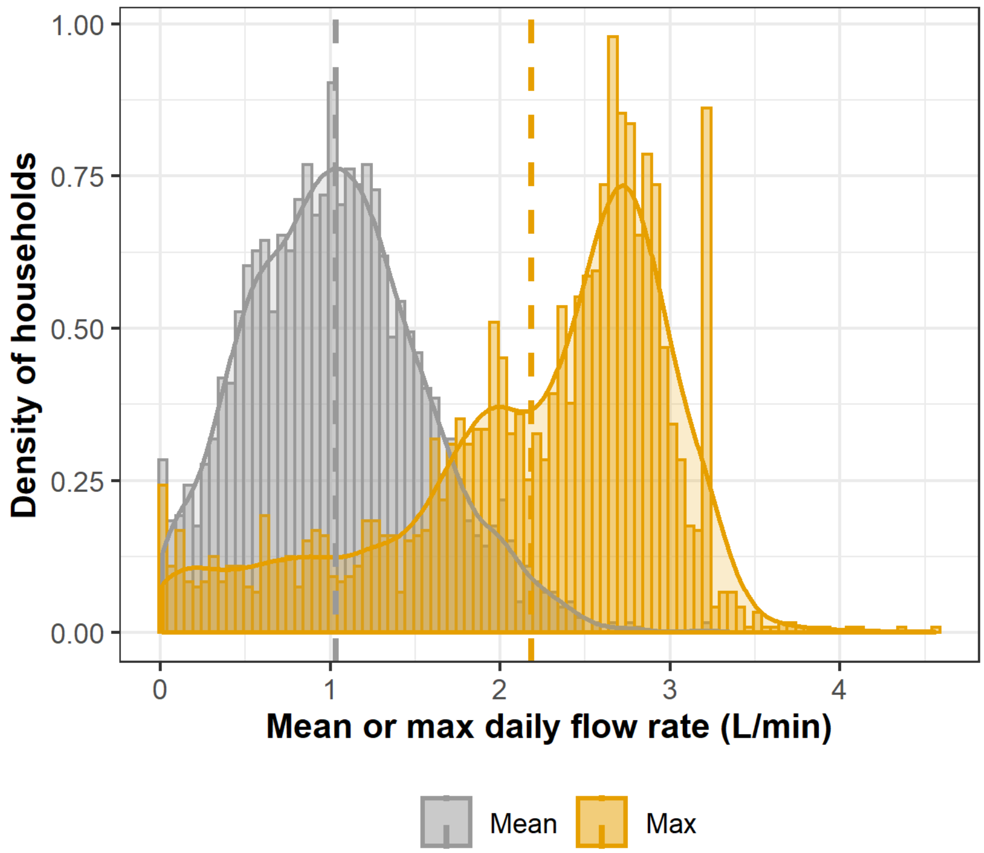

Figure A.2. Histogram of the daily mean and maximum flow rates among all participating households over the course of the study. Vertical dashed lines show the average mean and average max daily flow rates, respectively.

Table A.3. Range of removal performance observed for various metals for all households over the entire study period.

	Mean	*Med*	*Min.*	*Max.*
Al	4%	91%	-2213%	100%
As	50%	61%	-57%	97%
Cd	66%	85%	-171%	99%
Cu	90%	100%	-13%	100%
Fe	-43%	93%	-3873%	100%
Mn	-92%	-6%	-2326%	100%
Ni	14%	10%	-233%	96%
Pb	98%	100%	65%	100%
Sn	20%	0%	0%	99%
U	75%	93%	0%	99%
Zn	34%	93%	-1448%	100%

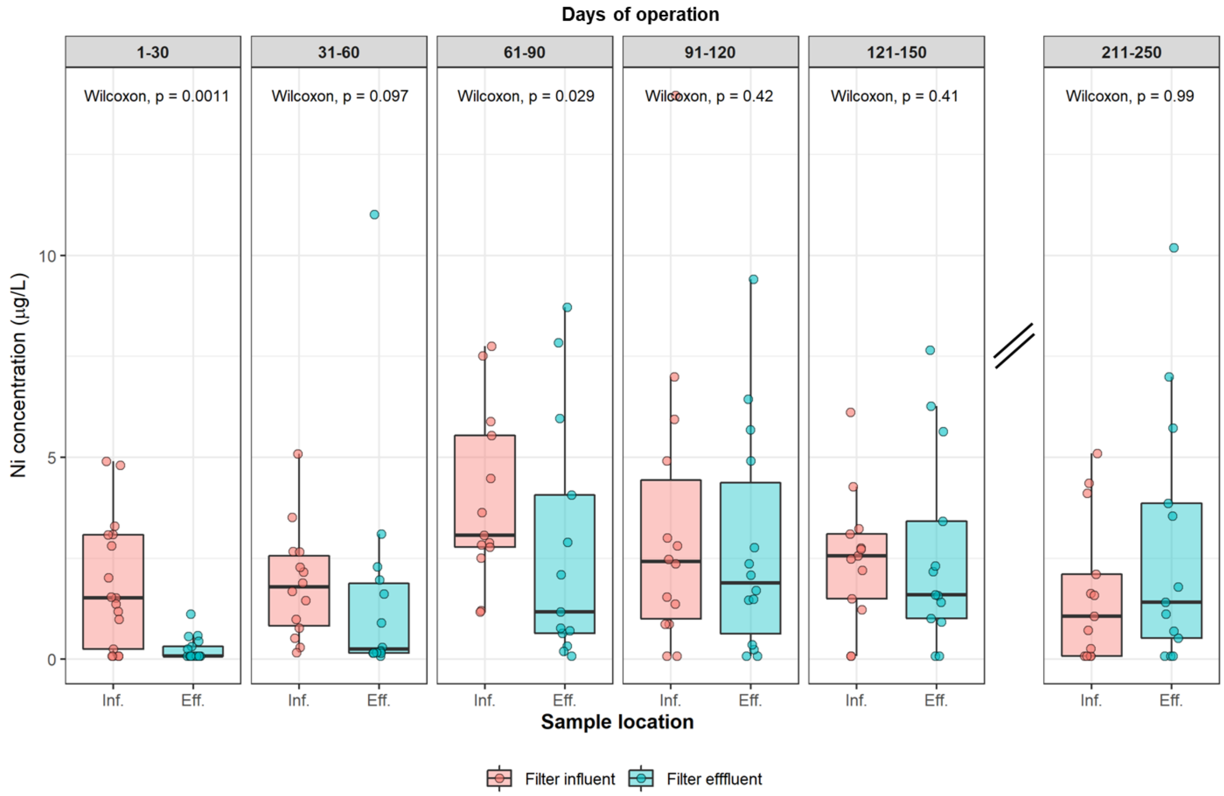

Figure A.3. Influent and effluent nickel concentrations over time showing median effluent concentrations approaching and surpassing median influent concentrations after approximately four months of use.

Table A.4. Pb results in baseline samples collected from each house (250 mL first draw without filter) compared to the average filter influent at each household during the study ± one standard deviation. All results in µg/L.

House ID	Baseline sample (250 mL first draw from faucet)	Average filter influent (1 L first draw from beneath the sink)
Cluster A		
5	2.72	2.24 ± 1.4
8	1.57	0.29 ± 0.1
9	0.13	0.13 ± 0.1
10	0.41	1.12 ± 0.8
Cluster B		
19	34.33	5.09 ± 3.6
21	0.40	0.42 ± 0.5
Cluster C		
1	9.93	8.36 ± 0.8
2	26.26	6.93 ± 2.6
3	7.99	4.13 ± 0.8
4	14.00	4.71 ± 0.7
7	0.69	3.45 ± 1.8
11	2.83	2.56 ± 0.9
13	5.47	1.28 ± 0.3
14	8.46	4.31 ± 1.4
15	20.27	4.42 ± 2.8
16	-	5.09 ± 3.6
17	9.36	4.90 ± 6.9

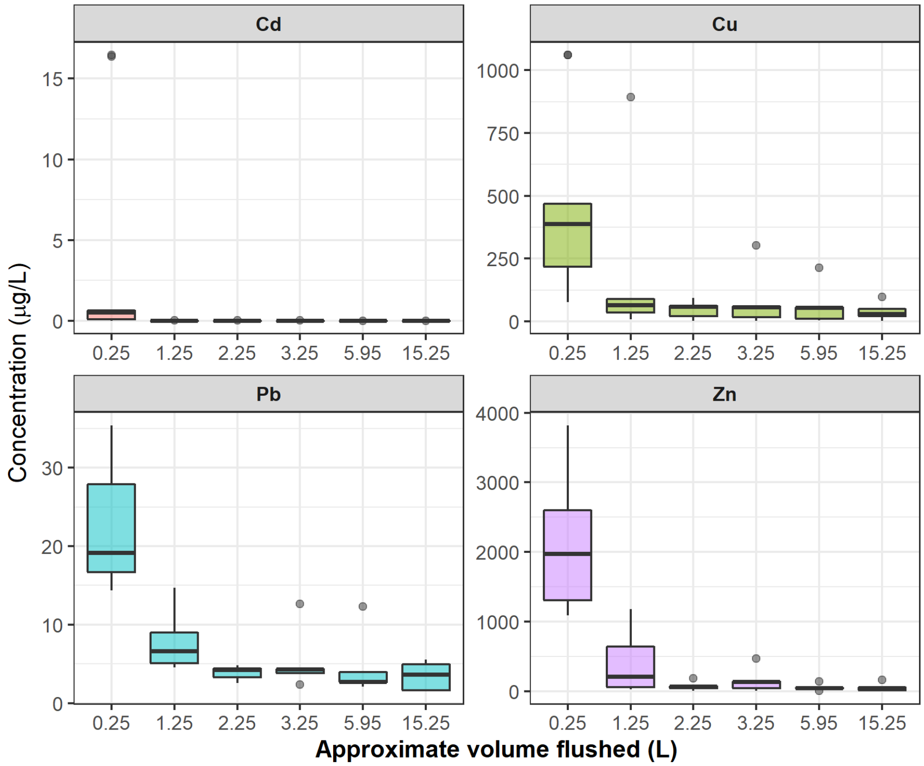

Figure A.4. Distribution of Cd, Cu, Pb, and Zn concentrations during first-draw sequential profile sampling in five homes before the filter was installed highlighting elevated concentrations in the first 250 mL of the profile due to interaction with the faucet fixture.

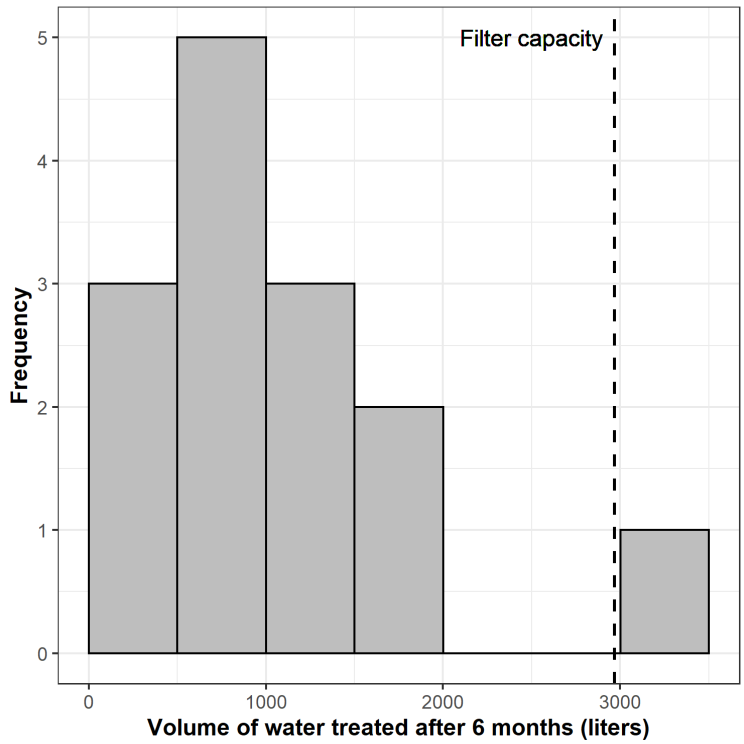

Figure A.5. Histogram of cumulative water usage after six months of use (excluding three households where the filter clogged before the six-month mark was reached).

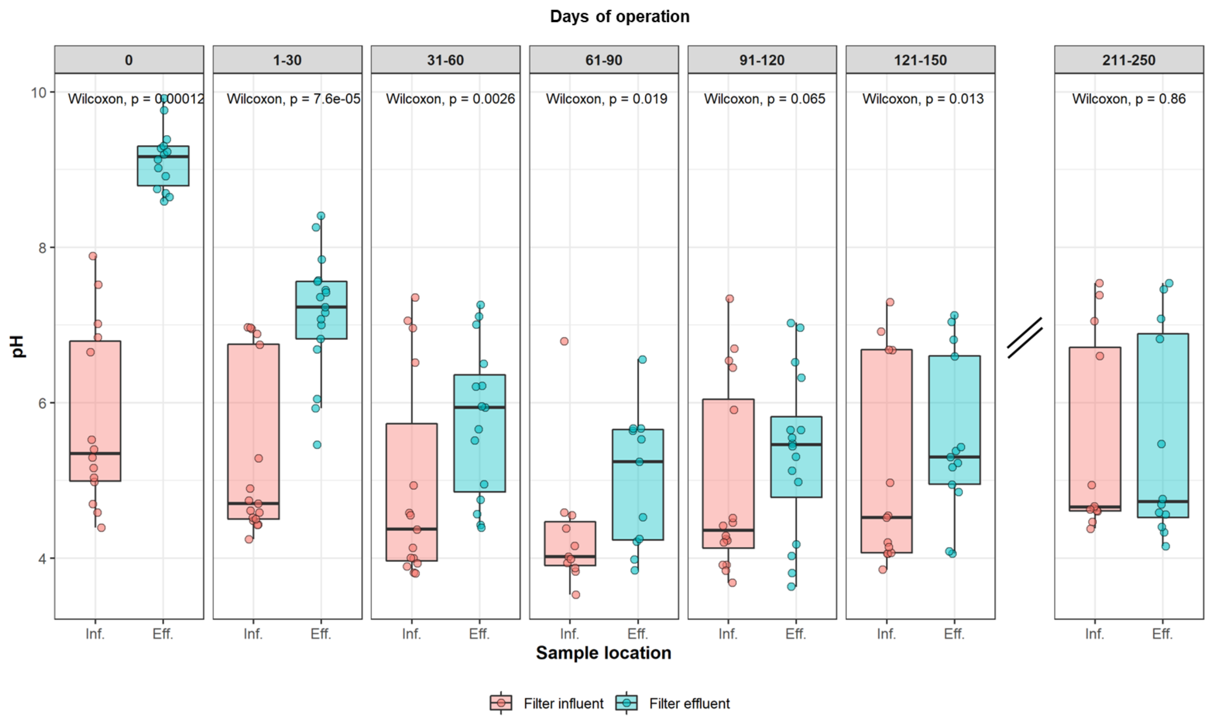

Figure A.6. Distribution of pH levels in the filter influent and effluent samples at each sampling month.

APPENDIX B: SUPPLEMENTARY MATERIAL FOR CHAPTER 3

Table B.1. Surface area and pore volume characterization of the carbon material used to produce the activated carbon block in the tested filter compared to other commercial powdered activated carbons (Mulhern et al. 2017).

	This study	**Evoqua AC1230C**	**Calgon F400**	**Cabot-Norit HD3000**
Carbon material	Coconut-shell	Coconut shell	Bituminous coal	Lignite coal
BET surface area (m^2/g)	444	1157	932	525
Micropore surface area (m^2/g)	399	1010	839	450
Micropore volume (cm^3/g)	0.16	0.44	0.33	0.15

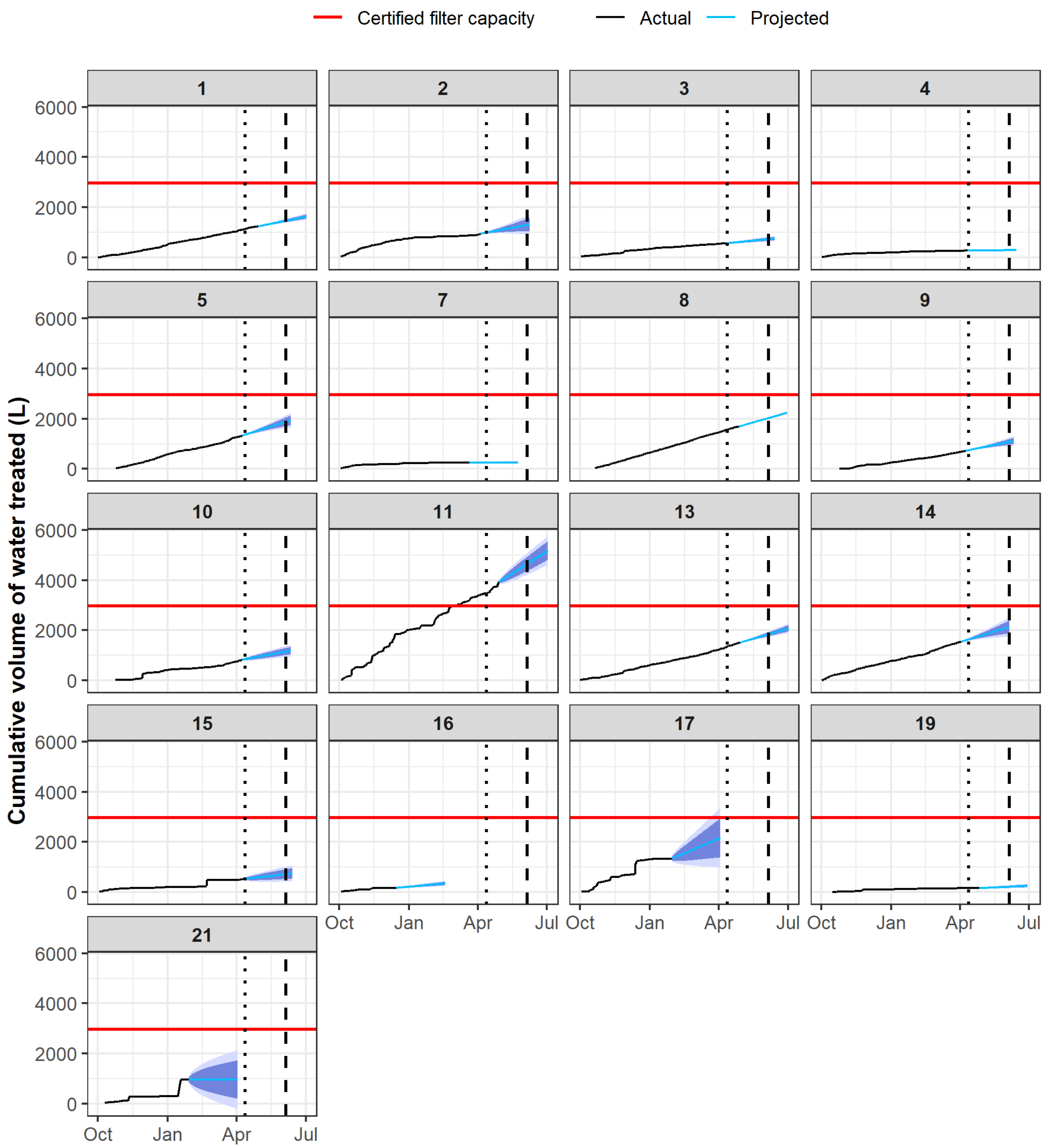

Figure B.1. Additive forecast model to estimate the cumulative volume of water treated by each household at the study end based on past usage patterns to account for missing sample months. Blue lines with confidence bands indicate forecast estimations. Horizontal red line shows the filter's recommended volume-based capacity (2967 L). Dotted vertical line indicates the approximate six-month mark since the filter was installed. Dashed vertical line indicates the study end date. The intersection of the blue forecast line and the vertical dashed line was used as the final estimate of the cumulative volume treated in each household at the end of the study.

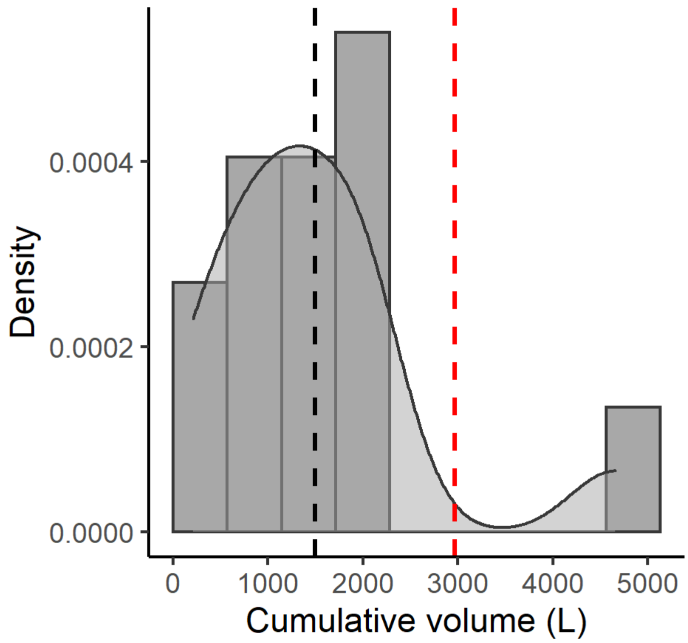

Figure B.2. Cumulative volume of water treated among all households after approximately eight months of use, not including three households where the filter clogged prematurely. Red vertical dashed line indicates the filter's recommended capacity (2967 L). Black vertical dashed line indicates the mean cumulative water use (1495 L).

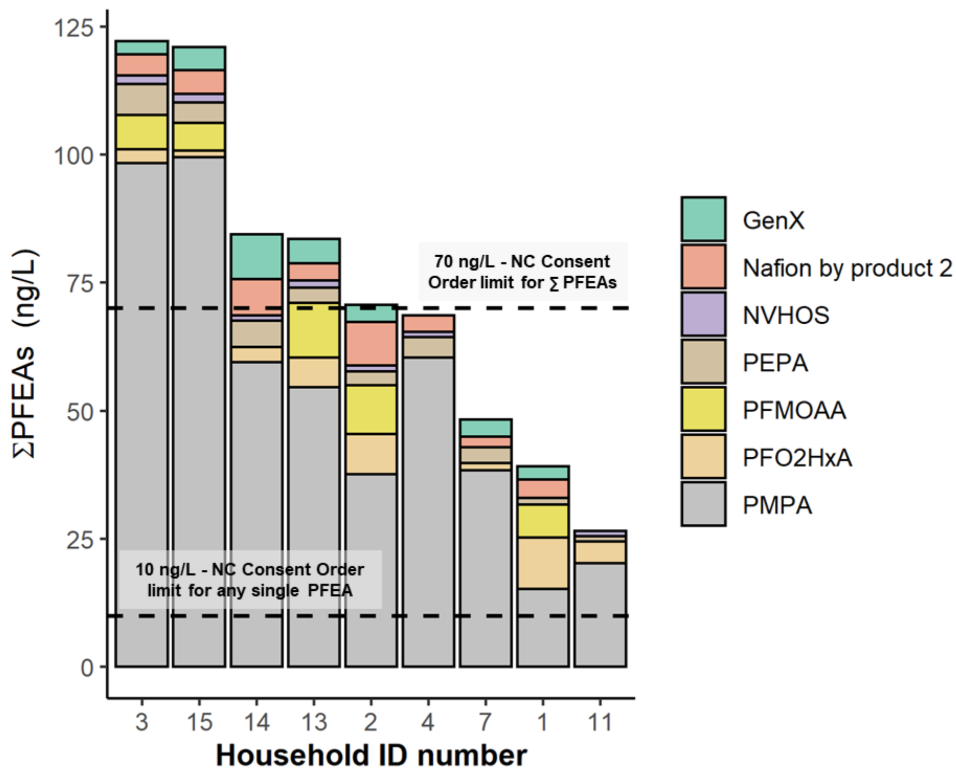

Figure B.3. Influent concentrations of PFEAs in households in cluster C near a fluorochemical manufacturing facility compared to the legal limits for PFEAs in drinking water supplies and private wells surrounding the facility set by a 2019 North Carolina Consent Order (North Carolina General Court of Justice 2019).

Table B.2. Mixed effects Tobit regression results predicting the effect of bed volumes, influent PFAS, DOC and pH on log(effluent) concentration.

Predictor		PFBA	PFBS	PFHpA	PFHxA	PFHxS	PFPeA	GenX, PFOA, PFOS, PFPeS
BV100	β	0.01	-0.15	-0.08	0.03	0.37	0.05	Insufficient or no effluent results >MDL
	p-value	0.85	0.11	0.67	0.74	0.21	0.18	
log(Influent PFAS)	β	0.19	0.54	1.03	1.40	-22.05	0.23	
	p-value	0.73	0.16	0.60	0.32	0.23	0.57	
Influent DOC	β	-7.4E-5	4.6E-4	-5.8E-04	-0.01	-0.05	0.00	
	p-value	0.94	0.45	0.82	0.43	0.26	0.41	
Influent pH	β	-0.45	-0.26	-1.08	0.77	-1.29	0.19	
	p-value	0.40	0.53	0.55	0.72	0.77	0.67	

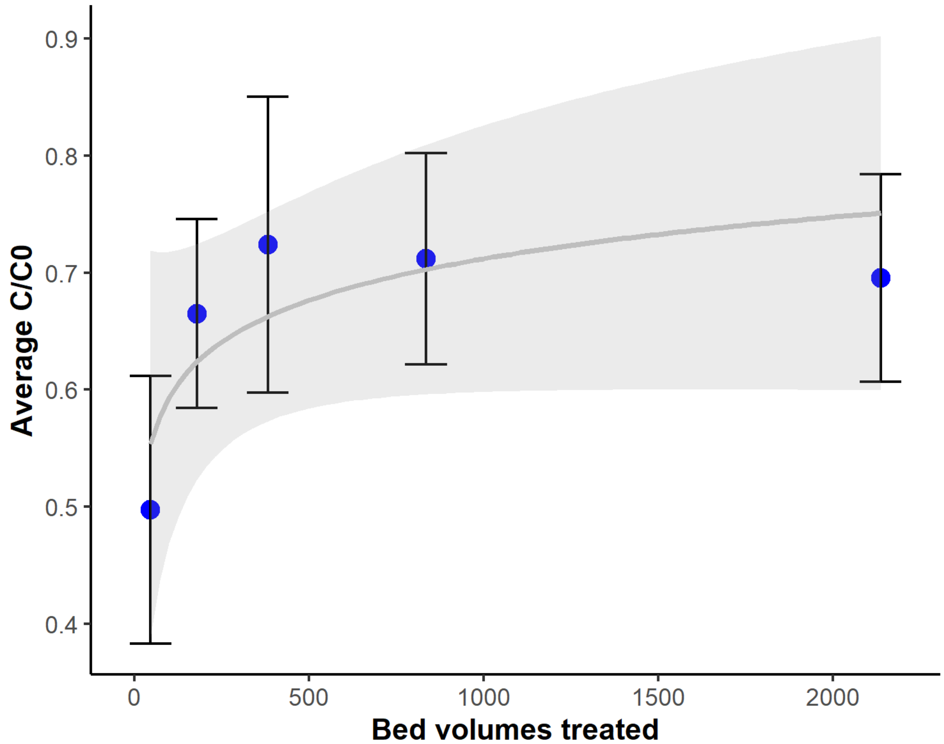

Figure B.4. Average DOC breakthrough aggregated by quintiles of bed volumes treated. Error bars show standard error around the mean DOC C/C_0 value among all households for each quintile.

Table B.3. Complete list of PFAS analytes included in both analytical methods. Method I used solid-phase extraction according to USEPA 533. Method II used large volume direction injections according to a method developed by North Carolina State University.

Analyte	Chemical formula	Chain length	CAS#	Chemical name	Method
Perfluorosulfonic acids (PFSAs)					
PFBS	$C_4HF_9O_3S$	4	375-73-5	Perfluorobutane sulfonic acid	I
PFPeS	$C_5HF_{11}O_3S$	5	2706-91-4	Perfluoropentane sulfonic acid	I
PFHxS	$C_6HF_{13}O_3S$	6	355-46-4	Perfluorohexane sulfonic acid	I
PFHpS	$C_7HF_{15}O_3S$	7	375-92-8	Perfluoroheptane sulfonic acid	I
PFOS	$C_8HF_{17}O_3S$	8	1763-23-1	Perfluorooctane sulfonic acid	I
PFNS	$C_9HF_{19}O_3S$	9	68259-12-1	Perfluorononane sulfonic acid	II
PFDS	$C_{10}HF_{21}O_3S$	10	335-77-3	Perfluorodecane sulfonic acid	II
Perfluorocarboxylic acids (PFCAs)					
PFBA	$C_4HF_7O_2$	4	375-22-4	Perfluorobutanoic acid	I
PFPeA	$C_5HF_9O_2$	5	2706-90-3	Perfluoropentanoic acid	I
PFHxA	$C_6HF_{11}O_2$	6	307-24-4	Perfluorohexanoic acid	I
PFHpA	$C_7HF_{13}O_2$	7	375-85-9	Perfluoroheptanoic acid	I
PFOA	$C_8HF_{15}O_2$	8	335-67-1	Perfluorooctanoic acid	I
PFNA	$C_9HF_{17}O_2$	9	375-95-1	Perfluorononanoic acid	I
PFDA	$C_{10}HF_{19}O_2$	10	335-76-2	Perfluorodecanoic acid	I
PFUnA	$C_{11}HF_{21}O_2$	11	2058-94-8	Perfluoroundecanoic acid	I
PFDoDA	$C_{12}HF_{23}O_2$	12	307-55-1	Perfluorododecanoic acid	I
PFTrDA	$C_{13}HF_{25}O_2$	13	72629-94-8	Perfluorotridecanoic acid	II
PFTeDA	$C_{14}HF_{27}O_2$	14	376-06-7	Perfluorotetradecanoic acid	II
Per- and polyfluoroalkyl ether acids (PFEAs)					
PFMOAA	$C_3HF_5O_3$	3	674-13-5	Difluoro(perfluoromethoxy)acetic acid	II
PFO2HxA	$C_4HF_7O_4$	4	39492-88-1	Perfluoro-3,5-dioxahexanoic acid	II
PMPA	$C_4HF_7O_3$	4	13140-29-9	Perfluoro-2-methoxypropanoic acid	II
PFMPA	$C_4HF_7O_3$	4	377-73-1	Perfluoro-3-methoxypropanoic acid	I
PFEESA	$C_4HF_9O_4S$	4	113507-82-7	Perfluoro(2-ethoxyethane)sulfonic acid	I
NVHOS	$C_4H_2F_8O_4S$	4	801209-99-4	1,1,2,2-Tetrafluoro-2-(1,2,2,2-tetrafluoroethoxy)ethanesulfonic acid	II
PEPA	$C_5HF_9O_3$	5	267239-61-2	Perfluoro-2-ethoxypropanoic acid	II
PFMBA	$C_5HF_9O_3$	5	863090-89-5	Perfluoro-4-methoxybutanoic acid	I
PFO3OA	$C_5HF_9O_5$	5	39492-89-2	Perfluoro-3,5,7-trioxaoctanoic acid	II
PFO4DA	$C_6HF_{11}O_6$	6	39492-90-5	Perfluoro-3,5,7,9-butaoxadecanoic acid	II
GenX	$C_6HF_{11}O_3$	6	13252-13-6	Perfluoro-2-propoxypropanoic acid	I

PFO5DoDA	C7HF13O7	7	39492-91-6	Perfluoro-3,5,7,9,11-pentaoxadodecanoic acid	II
Nafion byproduct 1		7	29311-67-9		II
Nafion byproduct 2	C7H2F14O5S	7	749836-20-2	Perfluoro-2-{[perfluoro-3-(perfluoroethoxy)-2-propanyl]oxy}ethanesulfonic acid	II
Nafion byproduct 4					II
ADONA	C7H2F12O4	7	919005-14-4	4,8-Dioxa-3H-perfluorononanoic acid	I
Hydro EVE acid	C8H2F14O4	8	773804-62-9		II
9ClPF3ONS	C8HClF16O4S	8	756426-58-1	9-Chlorohexadecafluoro-3-oxanonane-1-sulfonic acid	I
F53B Major	C8ClF16KO4S	8	73606-19-6	Potassium 9-chlorohexadecafluoro-3-oxanonane-1-sulfonate	II
F53B Minor	C10ClF20KO4S	10	83329-89-9	Potassium 11-chloroeicosafluoro-3-oxaundecane-1-sulfonate	II
11ClPF3OUdS	C10HClF20O4S	10	763051-92-9	11-Chloroeicosafluoro-3-oxaundecane-1-sulfonic acid	I
Fluorotelomer sulfonates					
42FTS	C6H5F9O3S	6	757124-72-4	1H,1H, 2H, 2H-Perfluorohexane sulfonic acid	I
62FTS	C8H5F13O3S	8	27619-97-2	1H,1H, 2H, 2H-Perfluorooctane sulfonic acid	I
82FTS	C10H5F17O3S	10	39108-34-4	1H,1H, 2H, 2H-Perfluorodecane sulfonic acid	I
			Perfluoroalkyl sulfonamides		
PFBSA	C4H2F9NO2S	4	30334-69-1	Perfluorobutanesulfonamide	II
PFHxSA	C6H2F13NO2S	6	41997-13-1	Perfluorohexanesulfonamide	II
PFOSA	C8H2F17NO2S	8	754-91-6	Perfluorooctanesulfonamide	II
nMeFOSAA	C11H6F17NO4S	11	2355-31-9	N-methylperfluorooctane sulfonamidoacetic acid	II
nEtFOSAA	C12H8F17NO4S	12	2991-50-6	N-ethylperfluorooctane sulfonamidoacetic acid	II

B.1 Method I: Solid-phase extraction by USEPA 533

B.1.1 PFAS sample preparation and extraction

Because samples had been stored in the 1 L HDPE bottles that they had been collected in for a minimum of 7 months (and up to 18 months for the baseline samples collected in July 2019), several modifications were made to USEPA 533. Before analysis, the pH of each sample was measured to ensure it was in the range 6-8 as specified in USEPA 533. Any samples that were out of range were buffered with 1 g/L HPLC-grade ammonium acetate (Fisher Scientific, catalog #A639-500) and the pH was checked again. All samples were within the appropriate pH range after adding ammonium acetate buffer. Then, the entire sample volume was poured into a separate, virgin 1 L polypropylene container triple-rinsed with reagent water as a holding vessel (WebstaurantStore Choice, item #127RD32COMBO). No significant background levels of PFASs were detected from the rinsed holding vessels. The original sample bottle was then rinsed with 10 mL of methanol to ensure that any PFAS material that had absorbed to the inside of the bottle was returned to solution before analysis. The entire sample volume was then returned to the HDPE sample bottle and mixed well. The 1 L polypropylene holding vessel was used again to weigh out 250 grams of water from the sample bottle. The exact mass was recorded for calculating the final sample concentration after extraction and analysis. A 50 µL aliquot of the primary dilution standard using isotope dilution analogues was added to the subsampled volume and mixed well.

PFASs were extracted from the 250 mL sample using a weak anion exchange, polymeric sorbent (Phenomenex catalog #8B-S038-HCH) according to I at UNC Chapel Hill. PFASs were then eluted off the solid phase sorbent with 10 mL of methanol plus 2% ammonium hydroxide into 15 mL polypropylene conical centrifuge tubes (Falcon item #352196) 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 (v/v) for a final concentration factor of 500. The isotope performance standards were added to the reconstituted extracts and vortexed to mix.

B.1.2 Analysis of sample extracts

Extracts were stored at room temperature before analysis. A 10 µL aliquot of each sample was injected onto an Agilent 1290 Infinity LC system (Agilent, Santa Clara, CA) using a Zorbax Eclipse Plus C18 Rapid Resolution HD analytical column (3 x 30mm, 1.8 micron, Agilent, Santa Clara, CA) and Zorbax SB-C18 delay column (4.6 x 50 mm, 3.5 micron, Agilent, Santa Clara, CA) at a flow rate of 0.4 mL/min and column temperature of 50°C. The mobile phase composition consisted of 20 mM ammonium acetate in 95:5 water:acetonitrile (A) and 10 mM ammonium acetate in 95:5 acetonitrile:water (B). The initial gradient was set to 80% A and 20% B and adjusted according to **Table B.4** for a total run time of 15 min 12 sec. The eluting compounds were then quantified on an Agilent 6490 triple quadrupole MS/MS instrument using the following conditions: gas temperature: 230°C; gas flow: 11 L/min; sheath gas temperature: 350°C; sheath gas flow rate: 12 L/min; nebulizer pressure: 25 psi; capillary voltage: 2000 V; nozzle voltage: 0 V; high pressure RF: 90 V; low pressure RF: 60 V.

Table B.4. Mobile phase gradient conditions used in Method I.

Time (min)	A (%)	B (%)
0	80	20
1	60	40
6	50	50
13	15	85
13.1	0	100
15.1	0	100
15.2	80	20

B.1.3 Internal calibration curve

An internal standard calibration curve was prepared neat (without extraction) in 80% methanol using the USEPA 533 Native Analyte Primary Dilution Standard (Wellington EPA-533PAR). The six-point calibration curve ranged 0.25–50 ng/mL, representing a concentration range in the original unextracted samples of 0.5–100 ng/L. Prepared calibrators were analyzed as above at the beginning of each sample batch and fit using a linear regression. In addition, two continuing calibration checks, prepared as above, were run with each batch at the low and high end of the calibration curve. A reagent water double blank was run through the system after the calibration curve before analyzing the sample batch. All calibrators and quality control samples were spiked with 50 µL of Isotope Dilution Standards (Wellington EPA-533ES) to yield 5 ng/mL and 20 ng/mL final concentration. Calibrators and quality controls were also spiked with 25 µL Isotope Performance Standards (Wellington EPA-533IS) to yield 5 ng/mL and 15 ng/mL final concentrations.

B.1.4 Quality Control

Precision and accuracy were demonstrated during each sample run following USEPA 533 guidelines using replicate laboratory fortified blanks (LFBs) spiked at known concentrations at varying levels of the calibration curve. The relative standard deviation (RSD) was required to be within +/- 20% for all method analytes (**Table B.5**). Precision for the analyte 6:2 FTS was out of range during method development and this analyte was subsequently dropped from the method. Recovery of LFBs must be within 70 to 130% (**Table B.6**). Laboratory water spiked with primary dilution standard was also extracted with each sample run to verify low background from the extraction process.

Table B.5. Summary of relative standard deviation (RSD) calculated from replicate laboratory fortified blanks at three concentration levels for all analytes in Method I. Average RSD must be below 20%. All analytes passed except for 6:2 FTS which was dropped from the method.

	Spike concentration (ng/L)			
Analyte	**5**	**10**	**20**	**Average**
11ClPF3OUdS	33.8	0.3	14.3	16.1
36OPFHpA	18.2	9.3	6.7	11.4
42FTS	5.0	2.6	4.1	3.9
62FTS	39.2	10.0	28.8	**26.0**
82FTS	24.6	9.7	43.5	25.9
9ClPF3ONS	12.2	13.6	1.1	9.0
ADONA	11.1	18.4	14.2	14.6
GenX	9.1	18.4	15.7	14.4
PFBA	1.3	9.9	3.6	4.9
PFBS	13.0	10.7	5.3	9.6
PFDA	4.4	11.2	6.0	7.2
PFDoDA	10.8	12.2	16.4	13.1
PFEESA	17.2	16.2	1.3	11.6
PFHpA	10.9	23.3	5.0	13.1
PFHpS	3.5	7.6	6.2	5.8
PFHxA	6.8	7.7	3.5	6.0
PFHxSK	8.0	15.9	2.2	8.7
PFMBA	19.4	14.9	8.2	14.2
PFMPA	21.1	23.0	13.4	19.1
PFNA	17.9	9.6	10.6	12.7
PFOA	23.0	11.9	10.1	15.0
PFOS	1.6	8.7	1.7	4.0
PFPeA	3.7	13.0	3.0	6.6
PFPeS	4.8	5.0	4.7	4.8
PFUnA	19.3	12.4	14.6	15.5

Table B.6. Mean recovery of all fortified laboratory blanks for all analytes in Method I. Recovery must be within 70–130%.

Analyte	Mean recovery
11ClPF3OUdS	93%
36OPFHpA	103%
42FTS	90%
62FTS	123%
82FTS	122%
9ClPF3ONS	89%
ADONA	76%
GenX	96%
PFBA	89%
PFBS	90%
PFDA	93%
PFDoDA	97%
PFEESA	76%
PFHpA	91%
PFHpS	96%
PFHxA	89%
PFHxSK	89%
PFMBA	90%
PFMPA	87%
PFNA	95%
PFOA	99%
PFOS	90%
PFPeA	87%
PFPeS	99%
PFUnA	100%

B.2 Method II: Large volume direct injection

B.2.1 UPLC-MS/MS Analysis

Prior to analysis, 900 µL of the sample was added to a 1.5 mL polypropylene microcentrifuge tube with 100 uL of internal standard. The tube was centrifuged at 15,000 rpm for 15 minutes at 4°C and 800 uL of the supernatant was transferred to a 2 mL polypropylene LCMS vial. A 200 µL aliquot of each sample was injected onto an Agilent 1290 LC system (Agilent, Santa Clara, CA) with 900 µL sample loop and separated on a Zorbax Eclipse Plus C18 analytical column (4.6 x 50 mm, 3.5 µ; Agilent, Santa Clara, CA). The mobile phase was comprised of 5 mM ammonium acetate in deionized water (A) and 5% deionized water and 5 mM ammonium acetate in HPLC-grade methanol (B). Gradient: initial 5% B to 95% B in 18 min, to 100% B in 4 min, for a total analysis time of 22 min. The column was re-equilibrated between each run for 6 min at initial conditions. Flow rate was 0.5 mL/min with a column temperature of 50°C. A 3 second needle wash consisting of Acetonitrile:Methanol: isopropyl alcohol:Water (v:v 1:1:1:1) was included before each injection to eliminate cross-contamination. The eluting compounds were then analyzed on an Agilent 6495c triple quadrupole mass spectrometry system (Agilent, Santa Clara, CA) using electrospray ionization in negative mode (ESI-) and in multiple reaction monitoring (MRM) mode. Due to the thermally labile nature of some PFAS analytes, two separate methods were run to quantitate the compounds with low source temperature or high source temperate settings with instrument parameters as follows: drying gas temperature: 100°C (low temperature method) or 290°C (high temperature method); drying gas flow rate: 15 L/min; sheath gas temperature: 250°C (low temperature method) or 350°C (high temperature method); sheath gas flow rate: 11 L/min; nebulizer pressure:15 psi; capillary voltage: 1500 V; nozzle voltage: 0 V; high pressure RF: 90 V; low pressure RF: 60 V.

The MRM transition list can be found in the supplemental table along with respective source temperature conditions.

B.2.2 Analyte Quantification

Analyte concentrations were calculated with an extracted internal standard calibration curve that was prepared by spiking calibration mix and labeled internal standard (IS) into DI water and analyzed with the previously described method and fitted with a quadratic regression and 1/x weight. The ten-point curve concentrations ranged from 1-500 ng/L and were analyzed at the beginning and end of the entire sample batch. In addition, three continuing calibration checks (ranging from 10-500 ng/L) were analyzed every 20 samples. The method reporting limit (MRL) was determined by selecting the lowest concentration calibration point that passed the requirements listed below and had a peak area that was 3x greater than the average method blank (neat solvent with IS) peak area. The method blanks were run in triplicate prior to the curve to obtain an average response. Calibration points were excluded from the curve if they did not pass ±30% accuracy, and 6 passing points were required with a 0.99 R2. Double blanks (neat solvent) were included before and after the calibration curve and each continuing calibration check and their concentration had to be ≤ ½ the MRL. See supplemental table for native and labeled IS pairing.

B.2.3 Quality Control Requirements

The following guidelines, obtained from DoD Quality Systems Manual Ver 5.3 (Department of Defense 2019), were used to ensure accurate analyte quantification: analyte retention time: ±0.4 min.; signal to noise ratio: ≥ 10 for ions used for quantification and must be ≥ 3 for ions used for confirmation; IS percent deviation: ±50%; and ion ratio: ±30%.

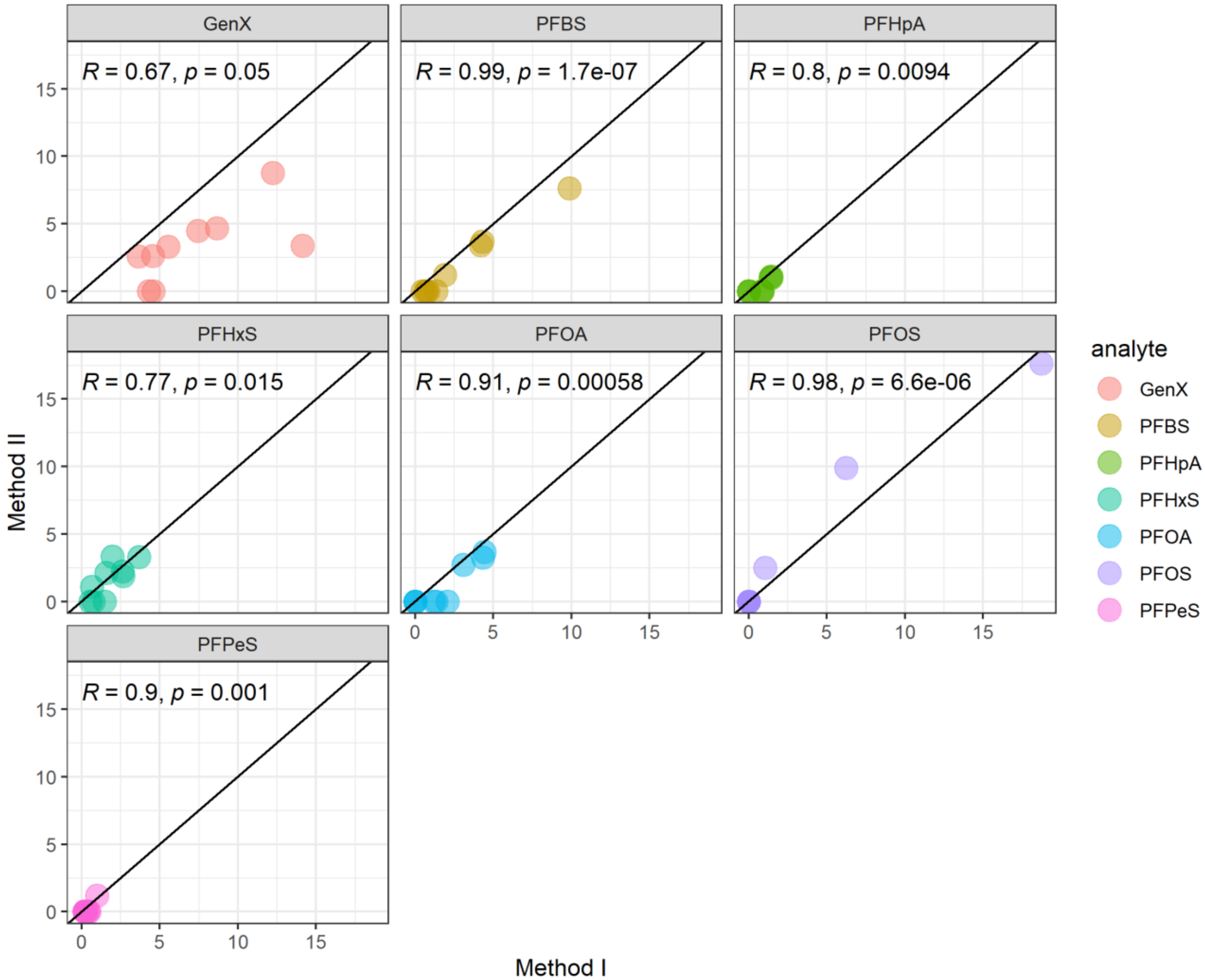

Figure B.5. Interlaboratory comparison for analytes included in both Method I and Method II for the nine influent samples analyzed under both methods showing high R values and statistically significant correlations, indicating good agreement between methods and laboratories.

APPENDIX C: SUPPLEMENTARY MATERIAL FOR CHAPTER 4

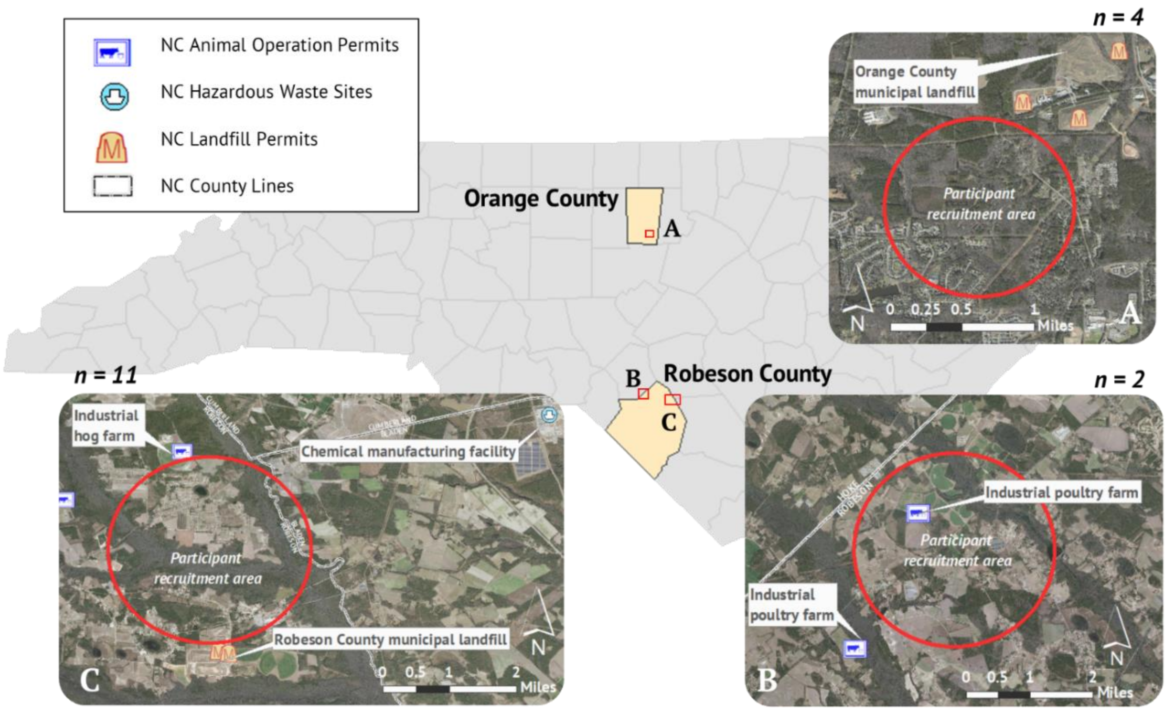

Figure C.1. Map of locations of study participants in North Carolina and surrounding environmental hazards.

Table C.1. Household-specific information of study participants within each geographic cluster.

Cluster	House ID	House type	Year of home construction	Year of well construction	Well depth (ft)	Distance from home septic system (ft)	Well upgradient?
A	5	Single family home	1985	Unknown	Unknown	115	Yes
	8	Single family home	1985	Unknown	Unknown	136	No
	9	Single family home	1987	1987	145	152	Yes
	10	Single family home	1990s	Unknown	Unknown	131	No
B	19	Single family home	1972	Unknown	25-30	97	Yes
	21	Single family home	1955	1955	26	38	No
C	1	Manufactured home/trailer	2010	Pre-2000	35	143	Yes
	2	Single family home	2000	2000	Unknown	64	No
	3	Manufactured home/trailer	1993	1996	35 ft	138	Yes
	4	Manufactured home/trailer	1990s	2000	35	92	Yes
	7*	Manufactured home/trailer	1976	Pre-1995	Unknown	124	Yes
	11	Manufactured home/trailer	2002	Unknown	Unknown	123	Yes
	13	Manufactured home/trailer	2018	2000	25-30	152	Yes
	14	Single family home	1998	2014	75	137	No
	15	Single family home	1997	Unknown	Unknown	94	Yes
	16*	Manufactured home/trailer	1970s	Pre-1995	Unknown	129	Yes
	17	Single family home	1983	1986	Unknown	152	Yes

*Two homes connected to the same well

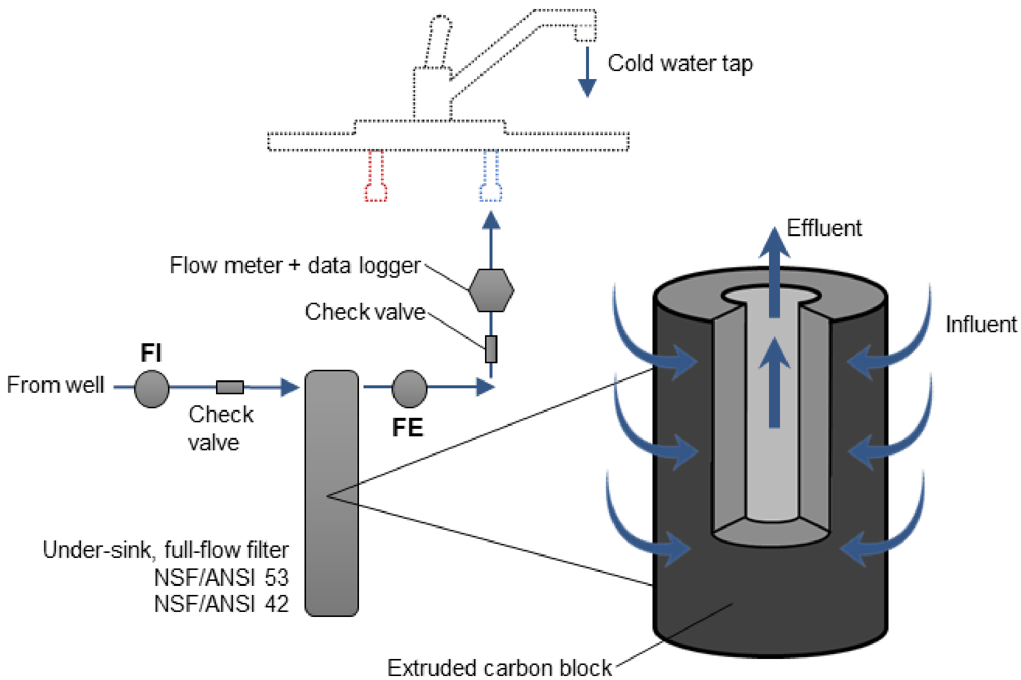

Figure C.2. Simplified schematic of POU filter installation beneath the primary kitchen sink. FI = filter influent; FE = filter effluent.

Table C.2. Complete list of predictor variables used during construction of multiple logistic regressions to identify significant predictors of microbial indicator organisms occurring in the filter effluent.

Variable name	Description
Binary outcome variables	
Total coliforms effluent	*Presence of total coliforms in the filter effluent* C_{eff} total coliforms >= 1 MPN/100 mL = 1 C_{eff} total coliforms <1 MPN/100 mL = 0
HPC increase effluent	*Increase of HPC in filter effluent* $HPC_{eff} > HPC_{inf} = 1$ $HPC_{eff} < HPC_{inf} = 0$
F+ coliphage effluent	*Presence of F+ coliphage in the filter effluent* C_{eff} F+ coliphage >= 1 PFU/100 mL = 1 C_{eff} F+ coliphage <1 PFU/100 mL = 0
Predictor variables	
Average daily time	Average amount of time the filter was in use per day (minutes) for the month prior to when the sample was taken
Average flow	Average flow rate of the filter (L/min) when the filter was being used during the month prior to when the sample was taken
Max flow	Maximum flow rate of the filter (L/min) when the filter was being used during the month prior to when the sample was taken
LPD	Average water usage (L/day) during the month prior to when the sample was taken
Cumulative volume	Overall water usage (L) for the entirety of the study from when the filter was installed to when the sample was taken
Cumulative volume – binary	Overall water usage (L) for the entirety of the study from when the filter was installed to when the sample was taken V > 50 L = 0 V < 50 L = 1
Cumulative time	Overall amount of time (weeks) the filter was in use from when it was installed to when the sample was taken
EC influent	Electrical conductivity (µS/cm) of the filter influent on the day the sample was taken
EC effluent	Electrical conductivity (µS/cm) of the filter effluent on the day the sample was taken
pH influent - binary	pH of the filter influent on the day the sample was taken pH >= 6 = 1 pH < 6 = 0
pH effluent	pH of the filter effluent on the day the sample was taken
Temp influent	Temperature of the filter influent on the day the sample was taken
Temp effluent	Temperature of the filter effluent on the day the sample was taken
Total coliform influent	Paired total coliform concentration (MPN/100 mL) in the filter influent at the time of the effluent sample
HPC influent	Paired heterotrophic plate count (CFU/mL) in the filter influent at the time of the effluent sample
F+ coliphage influent	Paired F+ coliphage concentration (PFU/100 mL) in the filter influent at the time of the effluent sample

Table C.3. Summary of influent and effluent microbial water quality in each geographic cluster.

		A (n=15)				**B (n=7)**				**C (n=44)**			
Sample location	**Analyte**	***mean***	***sd***	***range***	***% positive***	***mean***	***sd***	***range***	***% positive***	***mean***	***sd***	***range***	***% positive***
Influent	*pH*	6.9	0.3	6.5-7.4	-	5.0	1.3	3.8-6.9	-	4.3	0.4	3.5-5.3	-
	Temp (°C)	16	1	14-18	-	17	3	13-23	-	17	4	10-23	-
	Electrical Conductivity (µS/cm)	341	109	206-485	-	267	162	66-418	-	103	39	43-185	-
	HPC (CFU/mL)	5307	7780	20-25793	100%	536	752	65-2915	100%	353	1030	5-6660	73%
	Total Coliforms (MPN/100 mL)	1	2	1-7	7%	26	44	1-101	29%	1	1	1-4	7%
	F+ coliphage (PFU/100 mL)	4	5	1-16	73%	11	14	1-33	43%	5	7	1-23	48%
Effluent	*pH*	6.9	0.3	6.3-7.4	-	6.7	1.3	5.3-8.4	-	5.5	1.2	3.6-8.3	-
	Temperature (°C)	16	2	14-19	-	19	4	15-23	-	18	5	10-29	-
	Electrical Conductivity (µS/cm)	330	97	208-461	-	243	148	69-433	-	103	49	47-322	-
	HPC (CFU/mL)	863	803	55-2875	100%	853	994	5-2960	86%	956	1540	5-9760	98%
	Total Coliforms (MPN/100 mL)	1	0	1-1	7%	1	0	1-1	0%	59	334	1-2203	9%
	F+ coliphage (PFU/100 mL)	1	1	1-4	46%	6	12	1-30	43%	4	5	1-20	57%

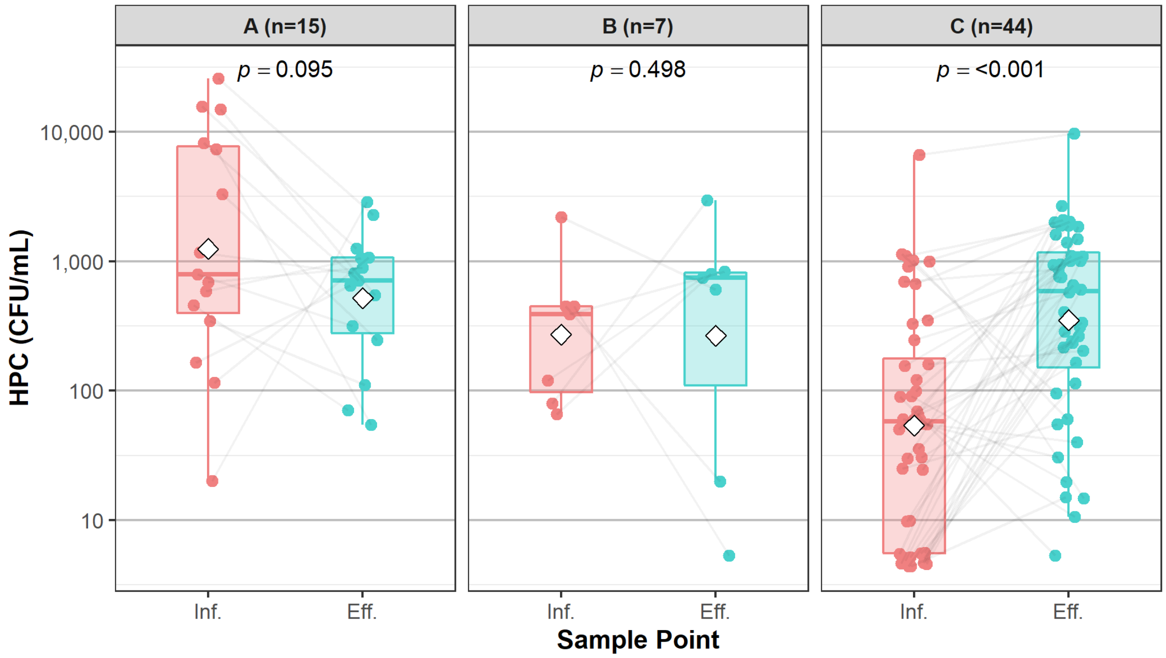

Figure C.3. Paired influent and effluent HPCs (CFU/mL) in each geographic cluster showing a significant increase in effluent HPC in cluster C but not in clusters A or B. *p* values represent the result of paired, non-parametric Wilcoxon signed rank tests.

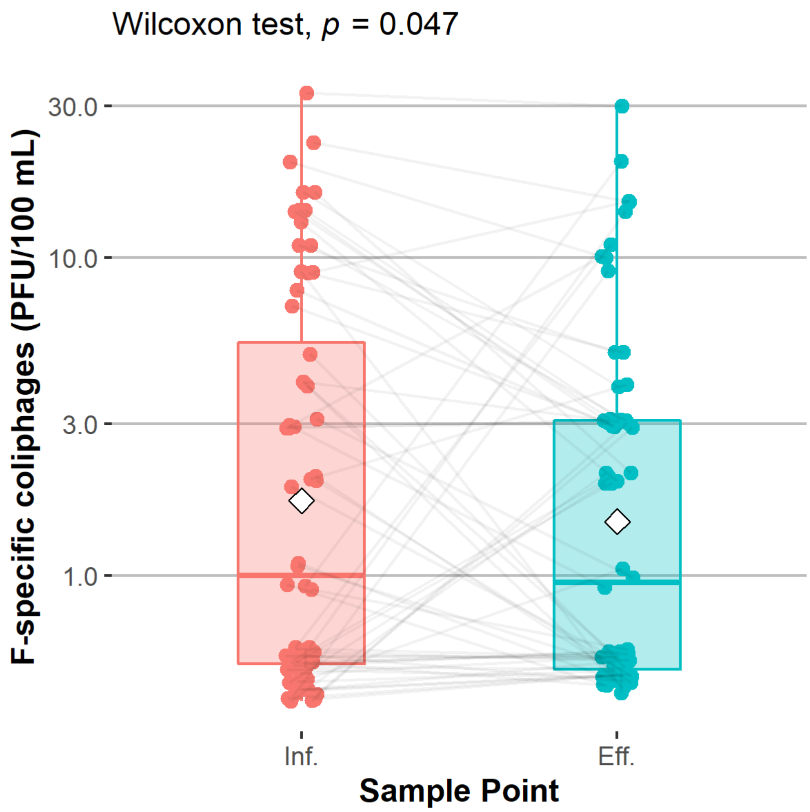

Figure C.4. Comparison of paired influent and effluent F-specific coliphage concentrations (PFU/100 mL) aggregated over the course of the study.

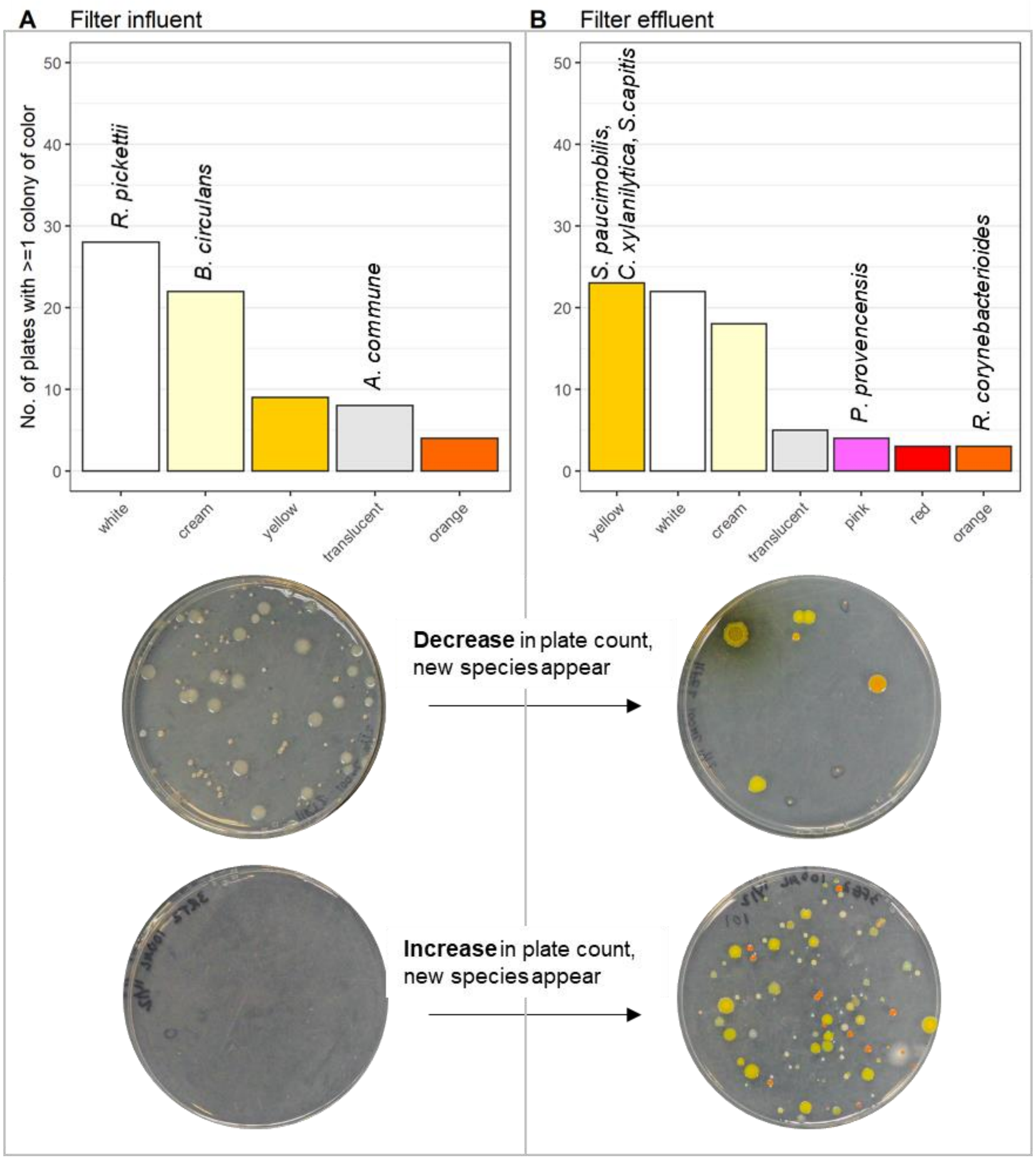

Figure C.5. Frequency of color descriptions of colonies on HPC plates in all filter influent and effluent samples over the course of the study. Increases in diversity (richness) were observed in cases where the overall effluent HPC both increased and decreased.

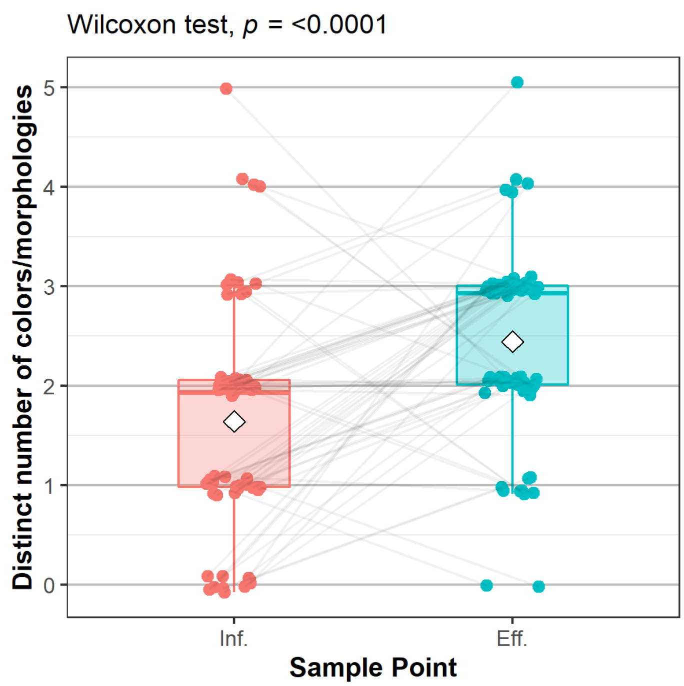

Figure C.6. Diversity of R2A plates for paired influent and effluent samples. A statistically significant increase in the median number of distinct colors and morphologies was detected in the effluent samples using paired Wilcoxon signed rank tests (p < 0.0001).

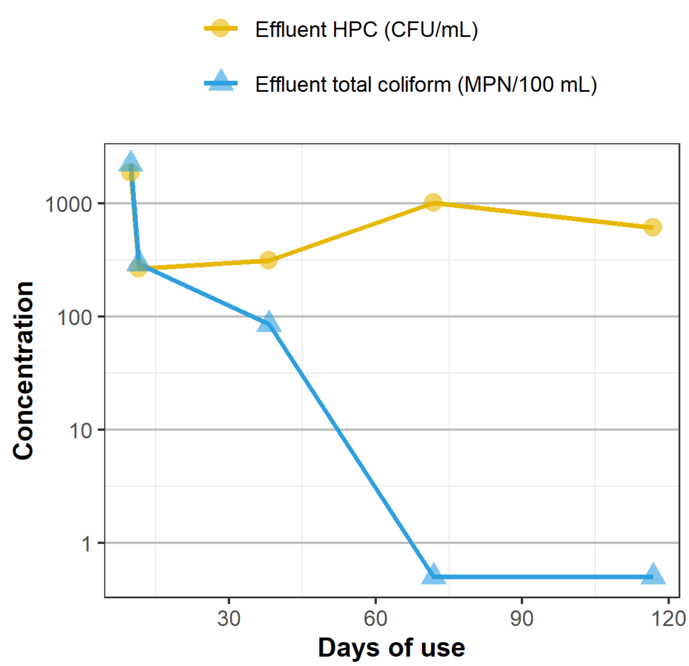

Figure C.7. Effluent concentrations of total coliform bacteria (CFU/100 mL) and HPC (CFU/mL) over four months of use from the filter in household #16.

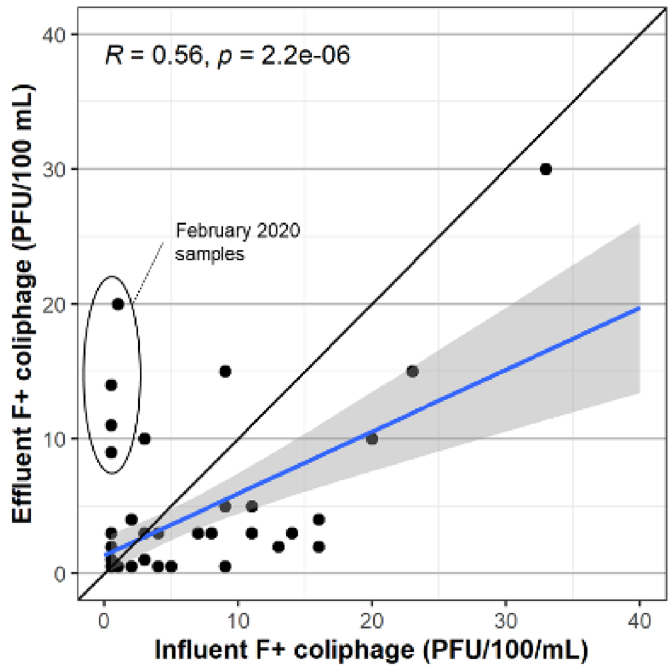

Figure C.8. A significant positive correlation exists between influent coliphage and effluent coliphage concentrations. February 2020 samples fall above the 1:1 line possibly due to shedding of viruses from the filter cartridge after periods of high influent concentrations.

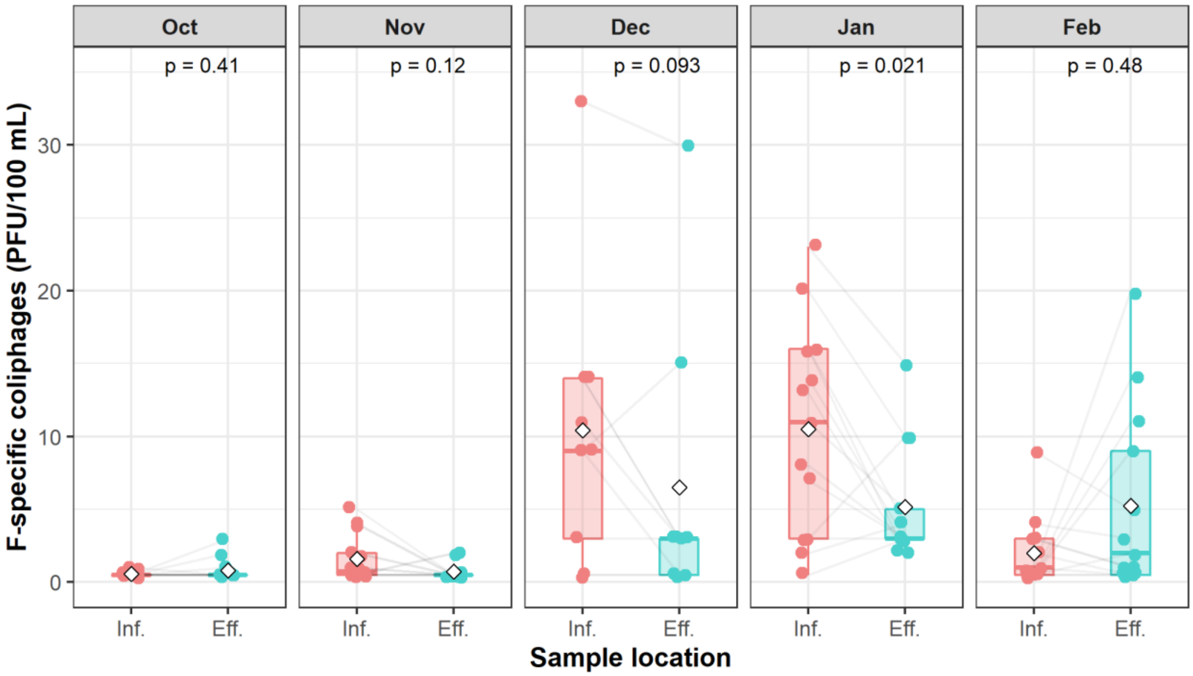

Figure C.9. Monthly paired influent-effluent F-specific coliphage concentrations reveal seasonal nature of coliphage concentrations in filter influent and possible viral shedding in effluent after influent concentrations subside.

DNA sequencing procedure and BLAST results

Taxonomical unites were identified for each bacterial isolate selected from TSA or R2A plates through 16S rRNA sequencing at a commercial laboratory (MR DNA, Shallowater, TX, USA). The amplification and sequencing method used are as follows.

The 16S rRNA gene was amplified using the HotStarTaq Plus Master Mix Kit (Qiagen, USA) with PCR primers 27F/1492R. A 35-cycle PCR was performed under the following conditions: 3 minutes at 94°C, followed by 35 cycles of 30 seconds at 94°C, 40 seconds at 53°C, and 90 seconds at 72°C. Lastly, a final elongation step was performed at 72°C for 5 minutes. After amplification, PCR products were evaluated for successful amplification in 2% agarose gel. Multiple samples were pooled together in equal proportions based on their molecular weight and DNA concentrations. The PCR pool was then purified using Ampure PB beads (Pacific Biosciences). Sequencing was performed on a PacBio Sequel instrument (Pacific Biosciences, Menlo Park, CA) according to the manufacturer's guidelines. After completion of initial DNA sequencing, each library underwent a secondary analysis and cleaning in which the sequencing data was depleted of barcodes, oriented 5' to 3', and sequences <150 base pair or ambiguous base calls were removed. Operational taxonomic units (OTUs) were defined by clustering at 3% divergence (97% similarity). This mapped sequence was then taxonomically classified using the curated Basic Local Alignment Search Tool (BLAST) (www.ncbi.nlm.nih.gov). The BLAST results for each sample are shown below.

Table C.4. Percent identity, e-values, and bit scores for bacterial isolates from water samples of filter influent and effluent.

Colony color on R2A	Morphology	Species identity	Percent identity	e-value	Bit score
Filter influent					
White	Circular	*Ralstonia pickettii*	100	0	2634
Tan/white	Circular	*Bacillus circulans*	78.8	0	1746
Translucent/ clear	Pinpoint circular	*Aquabacterium commune*	76.9	0	1535
Filter effluent					
Yellow glossy	Circular	*Sphingomonas paucimobilis*	97.9	0	2403
Yellow dull	Circular	*Staphylococcus capitis*	100	0	2657
Yellow pale	Circular	*Cellulomonas xylanilytica*	99.9	0	2599
Orange	Circular	*Rhodococcus corynebacterioides*	100	0	2596
Pink	Circular	*Paenibacillus provencensis*	99.9	0	2625

APPENDIX D: SUPPLEMENTARY MATERIAL FOR CHAPTER 5

Table D.1. Household-specific information of study participants within each geographic cluster.

Cluster	House ID	House type	Year of home construction	Year of well construction	Well depth (ft)
A	5	Single family home	1985	Unknown	Unknown
	8	Single family home	1985	Unknown	Unknown
	9	Single family home	1987	1987	145
	10	Single family home	1990s	Unknown	Unknown
B	19	Single family home	1972	Unknown	25-30
	21	Single family home	1955	1955	26
C	1	Manufactured home/trailer	2010	Pre-2000	35
	2	Single family home	2000	2000	Unknown
	3	Manufactured home/trailer	1993	1996	35 ft
	4	Manufactured home/trailer	1990s	2000	35
	7*	Manufactured home/trailer	1976	Pre-1995	Unknown
	11	Manufactured home/trailer	2002	Unknown	Unknown
	13	Manufactured home/trailer	2018	2000	25-30
	14	Single family home	1998	2014	75
	15	Single family home	1997	Unknown	Unknown
	16*	Manufactured home/trailer	1970s	Pre-1995	Unknown
	17	Single family home	1983	1986	Unknown

*Two homes connected to the same well

Table D.2. Factors and questions included in questionnaire delivered to study participants before and after participation in a six-month POU filter intervention. Questions with an asterisk were reverse coded in the scale sums.

Factor	Questions
Perceived vulnerability	• I drink my well water when I am at home.* • My well water is safe to drink.* • I feel comfortable drinking my well water.* • In the future I will drink my well water.* • My well water tastes funny. • My well water smells funny. • My well water comes out of the tap looking dirty. • In the future, I will drink my well water.*
Perceived benefits	• Treating my well water is important to my health. • Treating my well water is important to my family's health. • Using and maintaining a water filter in my home can protect me from harmful contaminants. • Water treatment isn't necessary for well water.*
Extended perceived benefits (post-test only)	All prior perceived benefits questions plus the following: • Household water filters can make my well water safe to drink. • Drinking bottled water is safer than using a water filter to treat my tap water.* • Buying water filters to treat my tap water can save me money in the long run. • My tap water tastes better since installing the filter. • My tap water smells better since installing the filter. • My tap water looks cleaner since installing the filter. • I trust my tap water more with the filter installed than before. • Household water filters work well for well water.
Perceived barriers (post-test only)	• Buying replacement water filters to treat my well water is too expensive for me. • Remembering to change out water filter cartridges is too difficult. • Buying bottled water to drink is cheaper than buying filters to treat my well water. • I am responsible for maintaining my own well water to make sure it is safe to drink.* • Household water filters are a practical solution for problems with my well water.*
Self-efficacy	*General:* • If my well water is contaminated, I can do something about it. *Research/knowledge acquisition:* • I can look up the recommended health limits for different chemicals in drinking water. • I can find someone to test my well water to make sure it is safe to drink. • I can find reliable information about problems with my well water. • I can find reliable information about how to treat my well water.

	POU treatment product selection/maintenance: • I can properly maintain a water filter for my home. • I can choose the correct type of water filter for my well water. *POU treatment implementation:* • I can do the plumbing to install a water filter at my kitchen sink.
Intent to purchase	*Treatment:* • In the future, I will purchase a replacement water filter for my kitchen sink. *Bottled water:* • In the future, I will buy bottled water to drink at home. *Testing:* • In the future, I will pay for my well water to be tested to make sure it is safe to drink.
Household water insecurity experiences scale (HWISE)	*Pre-test: In the last 4 weeks…* *Post-test: Since the filter was installed…* *…how frequently have you or anyone in your household experienced any of the following scenarios?* • You worried about your water. • Your main water supply was not available or not enough. • You could not wash your clothes because of problems with your water. • Problems with your water changed your schedule or plans. • Problems with your water changed what you ate. • You could not or did not want to wash your hands because of problems with your water. • You could not or did not want to bathe because of problems with your water. • There was no clean water to drink in your house. • You felt angry about your water situation. • You went to sleep thirsty. • There was no water in the house at all. • You felt ashamed because of problems with your water.
Race	*What race/ethnicity do you identify with?* • Black • White • American Indian or Alaskan Native • Asian • Pacific Islander • Hispanic/Latinx • Not listed
Education	*What is the highest level of education you have completed?* • Did not attend school • Less than high school

	• High school (through 12th grade) • Technical/vocational school • Associates degree • Some college, no degree • Bachelor's degree • Graduate degree
Household Income	*What was your household income last year before taxes?* • Less than \$20,000 • \$20,000–\$29,999 • \$30,000–\$39,999 • \$40,000–\$49,999 • \$50,000 or more

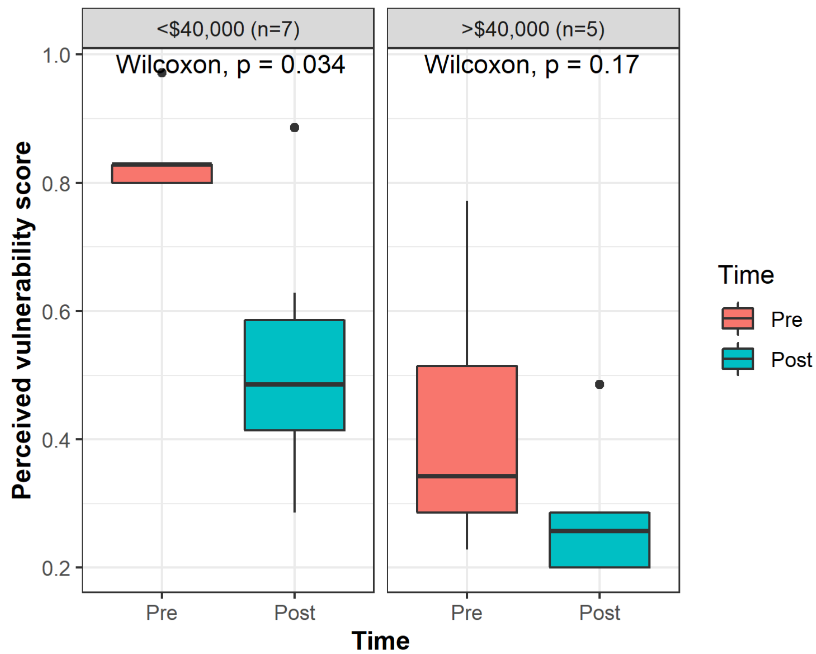

Figure D.1. Reduction in perceived vulnerability after the study among both low- and high-income groups.

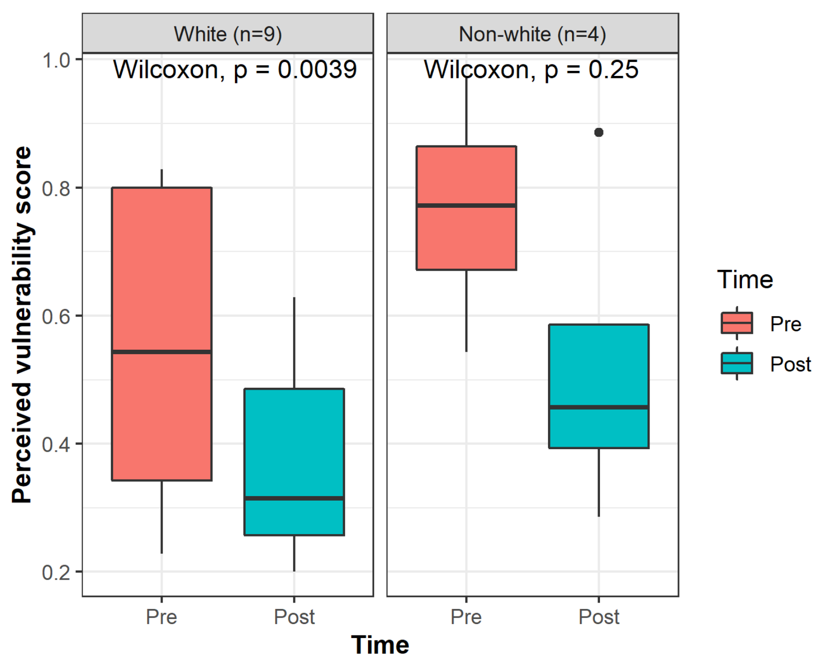

Figure D.2. Reduction in perceived vulnerability after the study among both white and non-white study participants.

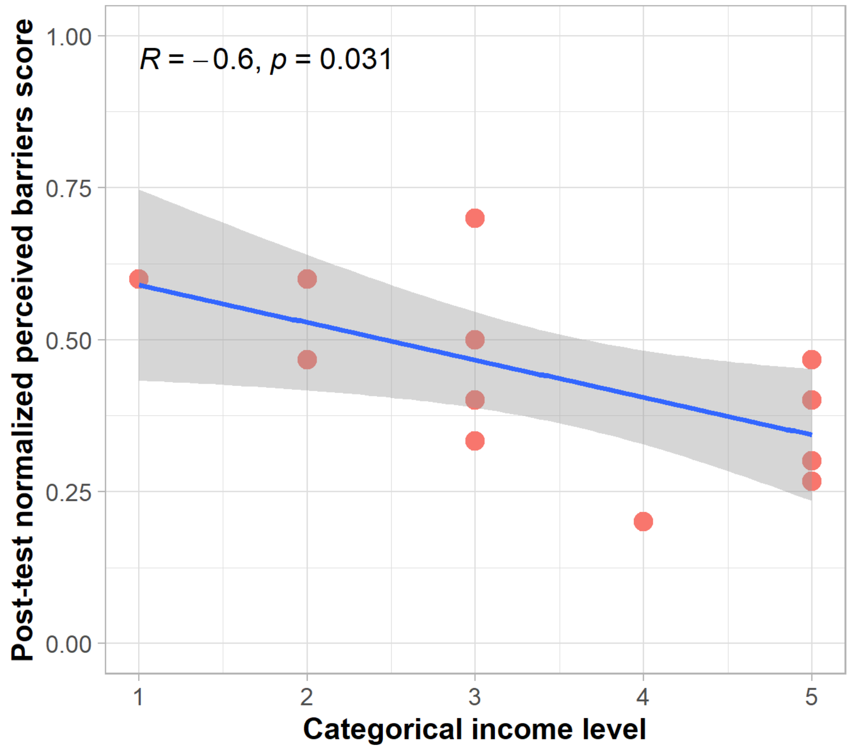

Figure D.3. Association between categorical income response and perceived barriers of POU water treatment after the study.