

**OCCURRENCE OF PER- AND POLYFLUOROALKYL SUBSTANCES (PFAS) IN
PRIVATE WATER SUPPLIES IN SOUTHWEST VIRGINIA**

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Occurrence of per-and polyfluoroalkyl substances (PFAS) in private water supplies in Southwest Virginia

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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are a class of man-made contaminants of increasing human health concern due to their resistance to degradation, widespread occurrence in the environment, bioaccumulation in human and animal organ tissue, and potential negative health impacts. Drinking water is suspected to be a primary source of human PFAS exposure, so the US Environmental Protection Agency (US EPA) has set interim and final health advisories for several PFAS species that are applicable to municipal water supplies. However, private drinking water supplies may be uniquely vulnerable to PFAS contamination, as these systems are not subject to EPA regulation and often include limited treatment prior to use for drinking or cooking. The goal of this study was to determine the incidence of PFAS contamination in private drinking water supplies in two counties in Southwest Virginia (Floyd and Roanoke), and to examine the potential for reliance on citizen-science based strategies for sample collection in subsequent broader sampling efforts. Samples for inorganic ions, bacteria, and PFAS analysis were collected on separate occasions by homeowners and experts at the home drinking water point of use (POU) in 10 Roanoke and 10 Floyd County homes for comparison. Experts also collected an outside tap PFAS sample. At least one PFAS compound was detected in 76% of POU samples collected (n=60), with an average total PFAS concentration of 23.5 parts per trillion (ppt). PFOA and PFOS, which are currently included in EPA health advisories, were detected in 13% and 22% of POU samples, respectively. Of the 31 PFAS species targeted, 15 were detected in at least one sample. On average, a single POU sample contained approximately 3 PFAS, and one sample contained as many as 8 different species, indicating that exposure to PFAS in complex mixtures is worth noting. Although there were significant differences in total PFAS concentrations between expert and homeowner collected samples (Wilcoxon, $\alpha = 0.05$), it is unclear whether this difference was due to contamination by the collector or the water usage and time of day of sampling (i.e. morning, afternoon). It is worth noting that there was no significant difference in the number of PFAS species in the samples collected by homeowners and experts. Given the considerable variation in PFAS detections between homes, future studies reliant on homeowner collection of samples appears possible given proper training and instruction to collect at the same time of day (i.e. first thing in the morning).

Occurrence of per-and polyfluoroalkyl substances (PFAS) in private water supplies in Southwest Virginia

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GENERAL AUDIENCE ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) belong to a large family of manmade compounds that are commonly used in a variety of household and consumer products due to their unique water and stain resistant properties. PFAS compounds are not easily broken down in the environment and have been detected globally in air, soil, and water samples. In addition to their environmental detections, PFAS are slow to be removed from the body after ingestion and known to cause negative health effects in concentrations less than one part per trillion. Drinking water is considered to a main source of PFAS consumption for humans; as such, the US Environmental Protection Agency (US EPA) has set strict, but not legally binding, interim and final health advisories (HA) for four types of PFAS. However, these health advisories only apply to public water services and do not cover private drinking water systems, such as wells or springs, which are the full responsibility of the well owner. Private drinking water system users often do not treat their water before drinking which may make these systems uniquely vulnerable to PFAS contamination. This study focused on 20 total homes, 10 in Roanoke County and 10 in Floyd County to see if PFAS was present and to determine whether or not homeowners would be able to collect their own samples for PFAS analysis at home as accurately as researchers or experts with proper instructions. Homeowners and experts collected drinking water samples inside at a point of use (POU), usually at a kitchen faucet, and outside of the home, usually from a tap. PFAS were present in 76% (n=60) of POU samples, with an average combined concentration of 23.5 parts per trillion (ppt). The two most well studied PFAS, PFOA and PFOS were detected in 13% and 22% of POU samples, respectively. It was also common to detect at least 3 PFAS in a single sample. Although there were differences in total average concentrations of PFAS in samples collected by homeowners and experts, variation could be caused by several factors indicating that with proper training and instruction it is likely future studies could still rely on homeowners to collect samples for PFAS analysis.

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1. Introduction: Per- and polyfluoroalkyl substances (PFAS) as an emerging contaminant in drinking water of increasing human health concern

Per and polyfluoroalkyl substances (PFAS) are part of a large group of anthropogenic contaminants of increasing concern to those in the environmental and public health spheres. PFAS species were originally developed to make a variety of consumer products resistant to water, grease, and stains. As a result of their unique chemical structure and hydrophobic nature, PFAS are mobile in various media including soil, water, and air, and are highly resistant to environmental degradation. Consequently, PFAS are commonly detected in groundwater and soil across the globe (Brusseau et al., 2020; Newell et al., 2021). Both long-chain perfluoroalkyl acids (PFCAs; $\geq 7C$ and PFSAAs; $\geq 6C$) and their short chain fluorinated alternatives such as PFBS and PFBA are associated with a variety of negative health effects (Fenton et al., 2021). Given frequent detection of PFAS in a variety of water sources, both treated and untreated, drinking water is currently suspected to be a major contributor of human exposure to PFAS compounds (Domingo & Nadal, 2019). Federal regulation and monitoring of PFAS in municipal drinking water supplies is rapidly evolving. In 2022, the US EPA issued final health advisories for PFBS and GenX, and interim health advisories for PFOA and PFOS, that are below the current level of analytical detection (Teymourian et al., 2021). However, it is worth noting that these advisories are not federally enforceable, and only apply to municipal drinking water treatment systems. The US EPA does not have jurisdiction over the estimated 23 million private drinking water wells in use nationally; monitoring for standard and emerging contaminants in these households' drinking water is the full responsibility of the homeowner (Murray et al., 2021). At present, the understanding of PFAS presence in private drinking water supplies is limited, and given minimal oversight and treatment, it is likely that these systems are uniquely vulnerable to contamination (D. Lee & Murphy, 2020)

As of January 2023, there is no nationally representative data set describing background PFAS concentrations in private well water (Hu et al., 2016; D. Lee & Murphy, 2020). Large-scale surveys of PFAS in groundwater have focused on source water levels (e.g., lakes and groundwater wells) rather than levels at the consumer point of use (McMahon et al., 2022). Limited field studies in North Carolina and Alaska have detected levels that exceed EPA's lifetime health advisory for total PFAS (70 ppt) though these studies specifically examined treatment efficacy (Mulhern et al., 2021) and the impact of a local rural airport (Babayev et al.,

2022). As most private systems do not typically employ treatment devices (Smith et al., 2014), and local land use can be diverse, full characterization of PFAS at the tap in private systems remains virtually unexplored.

1.1 Study Objectives

The overall goal of this study is to determine the incidence of PFAS contamination in private drinking water supplies in two counties in Southwest Virginia. This effort also aims to establish a standard homeowner sampling protocol that will be applied by researchers in larger future statewide PFAS investigations. Specifically, this paired study will address the following questions:

1. What species of PFAS are present in private drinking water supplies in Roanoke and Floyd Counties at the point of use (POU)?
2. Does homeowner collection of samples for subsequent PFAS analysis introduce bias when compared to expert collected samples?

This effort represents an important first step in quantifying PFAS concentrations in private drinking water supplies across the state, with significant implications for the ongoing management and regulation of emerging environmental contaminants in drinking water nationally. This work is likely of significant interest beyond Virginia and US borders as PFAS have been confirmed in groundwater as a source of municipal drinking water in virtually every modern nation in the world (Domingo and Nadal, 2019).

2. Literature Review

2.1 Per- and polyfluoroalkyl substances (PFAS) as emerging contaminants

2.1.1 A brief history of per-and polyfluoroalkyls substances (PFAS)

Per- and polyfluoroalkyl substances (PFAS, $C_nF_{(2n+1)}-R$) are a group of several thousand synthetic organic compounds initially engineered to meet diverse industrial and commercial needs. Since discovery of the unique chemistry of PFAS in the 1930s, this complex group of fluorinated organic compounds have become ubiquitous in consumer products, including cookware, cosmetics, and food packaging. An estimated 9,000+ PFAS and their degradants have been identified to date. PFAS molecules are identifiable by their unique chain of linked carbon atoms where the expected hydrogen atoms have been replaced with fluorine. Carbon-fluorine

bonds are one of the strongest in chemistry, making PFAS highly resistant to environmental and biological degradation (Wang et al., 2014). This strong bond confers a high level of chemical stability and surface tension lowering properties, creating stable foams, metal plantings, cleaning products, coatings, inks, and varnishes, among countless other products (Pervendouros et al., 2006). The stability and persistence of PFAS in hostile environments rendered them particularly useful in military, firefighting, aerospace, construction, and electronic industry applications (Kotthoff & Bücking, 2018; Sunderland et al., 2019)

However, both the widespread use and persistence of PFAS compounds has resulted in extensive inadvertent discharge to and accumulation within various environmental matrices. The discharge of PFAS from military and firefighting installations as well as unlined municipal landfills to surface and groundwater supplies is of particular concern (US EPA, 2021; Virginia Department of Health, 2021). Because PFAS accumulates in the environment, historical disposal and discharge sites can continue to contribute significant amounts of PFAS to soil and groundwater years after production has halted (Lindstrom et al., 2011). PFAS, particularly PFOA and PFOS, are now readily recoverable from environmental samples including finished drinking water, wastewater, wildlife tissues, and human blood serum (De Silva et al., 2021; USEPA 2022). The discharge of PFAS from military and firefighting installations as well as unlined municipal landfills to surface and groundwater supplies is of particular concern (Virginia Department of Health, 2021).

2.1.2 Per- and polyfluoroalkyl substance classifications

Estimates of the exact number of PFAS compounds now present in the global environment range from 4,000 to 9,000 distinct compounds (Inter-Organization Programme for the Sound Management of Chemicals, 2018; National Institute of Environmental Health Sciences, 2022). Given the breadth of chemical diversity within the broader group of PFAS, species are often grouped by their behavior, usage, and/or chemical structures. At the broadest level, the family of PFAS compounds consist of two major branches: non-polymers and polymers. Within the non-polymer family there are the two major groups of PFAS that are of the greatest interest to environmental and human health: per and polyfluoroalkyl substances. Perfluoroalkyl substances typically contain a functional head group, commonly a carboxylic or sulfonic acid, attached to a chain of fully fluorinated carbon atom-containing structures.

Polyfluoroalkyl substances, while similar in structure, have specific carbons in the structure that are not fluorinated. These two groups could be further broken down into a variety of subclasses including per fluorinated carboxylic acids, defluorinated phosphonic acids, and perfluoroalkyl phosphonic acids, each with unique and varied physiochemical properties (Wang et al., 2017).

The most structurally basic and commonly used PFAS are perfluoroalkyl acids (PFAAs). PFAAs can either be directly emitted to the environment or as the result of environmental degradation of polyfluoroalkyl substances in the environment and are subsequently the most widely studied subgroup of perfluoroalkyl substances. PFAA compounds are considered to be “terminal PFAS” or “terminal degradation products”, i.e. no further degradation products will form from them in the environment (ITRC, 2020). Within the PFAA family, compounds with eight or more carbons, with at least seven perfluorinated, are known as perfluoroalkyl carboxylic acids (PFCAs); while compounds with six or more carbons with at least six perfluorinated carbons, known as perfluoroalkane sulfonates (PFSAs) are considered “long chain”. Compounds of increasing regulatory concern including PFOS and PFOA are long chain members of the PFSA and PFCA families, respectively. While PFCAs and PFSAs have the capacity to contain up to twelve carbons, each family also contains a variety of compounds with less than seven carbons. PFCAs with seven or fewer carbons with six or less perfluorinated carbons or PFSAs with five or less carbons and five or less perfluorinated carbons are considered to be “short chains” (Brendel et al., 2018; Wang et al., 2017).

The creation of short chain or replacement PFAAs was driven by several factors including industry wide phase outs of long chain PFAAs, as well as mounting pressure from the Centers for Disease Control and Prevention in the early 2000s following initial concerns related to human health and exposure (Lewis et al., 2015). The phase out of long chain PFAAs resulted in the creation of novel short chains species such as perfluorobutane sulfonate (PFBS) and long chain precursors including the ammonium salt of hexafluoropropylene oxide dimer acid (HFPO-DA or GenX) to replace PFOA and PFOS. At the time of their introduction, short chain compounds were assumed to be a safer alternative to long chain PFAAs because they were less bioaccumulative in organ tissue (Brennan et al., 2021). Unfortunately, subsequent studies suggest that exposure to newer short chain PFAAs is still associated with negative effects on the kidneys, immune system, liver, reproductive system, and organ development (US EPA, 2022). While historical emissions of certain long chain PFAS also created unintended short chain by-

products, some short chain PFAS were already present in the environment prior to their commercial replacement (Wang et al., 2017). Although the use and emission of long chain PFAS, particularly polyfluorinated PFAS, has declined globally, a number of polyfluorinated substances can be partially degraded in the environment via a variety of biological and abiotic mechanisms (ITRC, 2022). The result is an increased frequency of detections of PFAAs in the environment that may yield unexpected temporal and spatial trends for areas without known PFAS producing entities. PFAAs have not been shown to further degrade or transform under standard environmental conditions. Short chain PFAS are still characterized by their C-F bond and so are extremely persistent in the environment, with higher mobility in soil and water than their long chain counterparts. Therefore, short chain PFAS can infiltrate more quickly to groundwater, potentially contaminating drinking water resources (Brendel et al., 2018). In addition to high mobility and persistence in the environment, short chain PFAS are also more difficult to remove from water sources through traditional absorption treatment technologies such as granular activated carbon (Li et al., 2020).

2.2 Per- and polyfluoroalkyl substances (PFAS) as a human health risk

2.2.1 Pathways for human exposure to per- and polyfluoroalkyl substances

Human exposure to PFAS can occur via multiple direct and indirect pathways, though at present, ingestion of drinking water and ingestion of contaminated food are assumed to be the primary contributors to observations of elevated human blood concentrations (Siebenaler et al., 2017). Multiple human health studies have confirmed that essentially all people living in industrialized countries have been exposed to PFAS compounds (Calafat et al., 2007; Jian et al., 2018; Kato et al., 2011; Khalil et al., 2018; Stubbleski et al., 2017), with detectable concentrations of PFOS and PFOA ranging from 3 to 29 ng/mL in blood samples (Kannan et al., 2004). Epidemiological studies have also revealed correlations between exposure to specific PFAS and a variety of negative health outcomes. Exposure to PFAS and/or PFAS biomarkers are associated with reduced immune and thyroid function (DeWitt et al., 2019; Grandjean et al., 2012; J. E. Lee & Choi, 2017), liver disease (Nian et al., 2022), lipid and insulin dysregulation (Andersson et al., 2019; Ballesteros et al., 2017; Blake et al., 2022; Caron-Beaudoin et al., 2019; Li et al., 2020), kidney disease (Barry et al., 2013; Shearer et al., 2021), adverse reproductive and developmental outcomes (Lopez-Espinosa et al., 2016), and various cancers (Steenland et al., 2020)

The current identified primary non-occupational human exposure route of concern is the ingestion of PFAS via public water supplies (Herkert et al., 2020). The first statewide study of PFAS in public US drinking water took place in New Jersey and reported detectable PFOA in 59% of public water distribution systems with reported maximum concentrations of nearly 200 ng/L (Post et al., 2009). Current estimates suggest that 18-80 million people in the United States receive treated drinking water with 10 ppt or greater total concentrations of PFOA and PFOS (Andrews & Naidenko, 2020). Confirmation of human PFAS uptake via waterborne ingestion has been confirmed in several field-based studies. West Virginia and Pennsylvania concluded that observations of elevated blood levels in local populations may be linked with past contamination of public drinking water systems (Pennsylvania Department of Health, 2019, West Virginia Department of Health, 2022). Pennsylvania found detectable PFOA in 100% of blood serum samples collected from 235 individuals living in Montgomery and Bucks County, counties classified as “high risk” for PFAS contamination due to the presence of military installations that used Aqueous Film-Forming Foams (AFFFs) for several decades. Water testing results from the highly publicized DuPont spill in the mid-Ohio Valley Region confirmed that drinking water in six water districts of two states (Ohio and West Virginia) were contaminated by PFOA as a result of chemical facility discharges into surface water (Bartell et al., 2010). Although the most detailed examinations of PFAS drinking water exposure have generally occurred near sites of past known industrial accidents, PFAS contamination of source waters appears quite widespread nationally (McMahon et al., 2022).

2.3 Federal regulation of per-and polyfluoroalkyls as a drinking water contaminant

2.3.1 PFAS under the Safe Drinking Water Act (SDWA)

Within the United States, the Safe Drinking Water Act (SDWA) requires the Environmental Protection Agency (US EPA) to address contaminants in public drinking water supplies and sources, including the development of enforceable standards and monitoring requirements for newly recognized or emerging contaminants (US EPA, 2022). The law protects both the finished drinking water that is ready for distribution to communities as well as sources of drinking water from groundwater and surface water (US EPA, 2022). The SDWA authorizes the US EPA to set national health-based standards that protect against both naturally occurring and anthropogenic pollutants that could contaminate drinking water (US EPA, 2022). These National Primary Drinking Water Regulations (NPDWR) are legally binding standards for public

water systems. The NPDWR set maximum contaminant levels (MCLs) for contaminants in drinking water that are a known public health threat such as lead and arsenic. Specific treatment techniques (TT) for removing these contaminants from water have also been established by the US EPA. Established standards also define secondary maximum contaminant levels (SMCLs) for contaminants that may cause aesthetic issues (e.g. drinking water taste, color, and odor) such as iron, but are not legally enforceable (US EPA, 2004). The US EPA can also establish a Maximum Contaminant Level Goal (MCLG), which describes a maximum level of a contamination in drinking water where no known or anticipated negative health effects occur within an established margin of safety, but these are also non-enforceable (US EPA, 2022).

To date, the US EPA's efforts to control adverse human and ecological exposures to PFAS have primarily focused on long chain, legacy species including PFOS and PFOA. Though virtually phased out of US production by major industry leaders for several years, these remain a critical legacy contamination concern due to high persistence (US EPA, 2021). In 2016, the US EPA established nonregulatory drinking water health advisories of 70 ppt for individual and total concentrations of perfluorooctanoic acid (PFOA) and perfluorooctanoic sulfonate (PFOS), the two most thoroughly studied PFAS (Post, 2020). A health advisory looks to protect all people, including high risk populations and life stages, from negative health effects resulting from a lifetime of exposure to various chemicals in drinking water, although health advisories can be issued for various exposure durations (e.g. 1 day, 10 days). Health advisories are not legally enforceable standards; rather, they are values developed to assist federal, state, tribal, and local officials in their efforts to protect public water resources especially in the case of a rapidly evolving situation (US EPA, 2021). The US EPA published lifetime health advisories for perfluorooctanoic acid (PFOA) and perfluorooctanoic sulfonic acid (PFOS) as part of the Third Unregulated Contaminant Monitoring Rule (UCMR3) in January 2017. Several US states, particularly states with former or current PFAS-producing conglomerates such as New Jersey, Minnesota, and California, subsequently concluded that these US EPA health advisories were insufficient. Since May 2020, nine states have developed drinking water standards or guideline values stricter than those initially issued by the US EPA, including concentration levels as low as 5.1 ppt (Post, 2021).

In 2021, the US EPA published the *PFAS Action Plan* which outlined potential steps to reduce PFAS in public drinking water. The Action Plan also included a call to set a federally

enforceable MCL under SDWA for various PFAS species, particularly PFOA and PFOS, by the end of 2022. In addition, the US EPA has proposed designating PFOA and PFOS, including their salts and structural isomers, to be regulated under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) or Superfund (US EPA, 2022). A hazardous substance designation would allow the US EPA to improve transparency between companies, local and state governments, and the public, hold companies liable for any environmental contamination resulting from PFAS, and designate companies to report releases of PFOA and PFOS (US EPA, 2022). Most recently, the US EPA published the Contaminant Candidate List 5 which included 18 additional PFAS compounds (US EPA, 2022). This report also included the first attempt to define and regulate PFAS as a class of contaminants rather than as individual species.

2.3.2 US EPA Established Health Advisories for PFAS

In June 2022, the US EPA announced new drinking water lifetime health advisories for four total PFAS compounds: interim health advisories (IHA) of 0.004 and 0.02 ppt for PFOA and PFOS, respectively, and final health advisories (FHA) of 10 ppt for GenX compounds (HPFO-DA and its ammonium salt) and 2000 ppt for PFBS (US EPA, 2022) (Table 1). A lifetime health advisory (LHA) as set by the US EPA identifies maximum permissible contaminant levels designed to protect all people, including sensitive populations such as immunocompromised individuals and children, from adverse health effects from exposure to PFAS in drinking water (US EPA, 2022).

Species	2016 Health Advisory	2022 Health Advisory
PFOA	≥ 70 ppt (Σ PFOA + PFOS or individual sample) *	0.004 ppt *
PFOS	≥ 70 ppt (Σ PFOA + PFOS or individual sample) *	0.02 ppt *
HFPO (Gen-X)	N/A	10 ppt **

PFBS	N/A	2,000 ppt **
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Table 1. US EPA non-enforceable health advisories set as of January 2023 for four PFAS compounds.

These health advisories are non-enforceable, non-regulatory guidelines for drinking water utilities and public health officials to use as a reference until MCLs are issued. As the new interim HA levels for PFOA and PFOS are currently below the level of detection and quantification for laboratory equipment (Teymourian et al., 2021), any detection of PFOA and PFOS in a sample, though not federally enforceable as of January 2023, is technically in exceedance of the interim lifetime health advisory (US EPA, 2022; Virginia Department of Health, 2022). Results of published toxicity studies are available for 31 PFAS of US EPA interest and assessment is underway for PFBA, PFHxS, PFHxA, PFDA, and PFNA (US EPA, 2022, Appendix A). The MCLs for PFOA and PFOS are still undergoing final approval and are now expected in 2023 (US EPA, 2022).

2.4 Private Drinking Water Systems

Although the widespread detection of PFAS in public drinking water supplies has garnered considerable national attention and more US EPA Health Advisories are expected in the coming years, it is critical to note that these regulations do not apply to private drinking water well systems, particularly wells, springs, and cisterns, which are the sole responsibility of the homeowner. A private water well is defined in the Code of Virginia as any water well constructed for a person on land which is owned or leased by that person and is usually intended for household, ground water source heat pump, agricultural use, industrial use or other nonpublic water well (Code of Virginia, 2009). Further, private systems serve fewer than 15 service connections or less than 25 people for at least 60 days a year. Because private systems can serve more than one household, the implications of a lack of treatment and system management may negatively affect an entire community. Many private wells do not include filtration, disinfection, or other treatments that target removal of common environmental contaminants including bacteria and metals prior to use or ingestion, making these systems uniquely vulnerable to contamination (D. Lee & Murphy, 2020; Smith et al., 2014). In a study of 2,100 domestic wells in

the US, water pumped from about 1 in 5 wells contained one or more contaminants (i.e. fecal indicator bacteria, nutrients, and metals) at a concentration greater than the SDWA established human-health benchmark for drinking water (DeSimone, 2009; Pieper et al., 2015). Health risks associated with private well water contamination is therefore of increasing concern. Recent work has connected fecal indicator bacteria positive wells and chronic homeowner gastrointestinal illness (Gruber et al., 2014), and documented higher levels of blood lead levels in children from communities served by private wells than those served by municipal water (Gibson et al., 2020).

2.4.1 PFAS detection and quantification in private systems in the US

Information on PFAS detection in private wells, specifically exposure at the tap, is very limited. As of January 2023, no nationally representative data set exists estimating existing PFAS concentrations in private well water (Hu et al., 2016; D. Lee & Murphy, 2020). The most complete study examining PFAS in groundwater used as a source for treated drinking water was completed by McMahon et al. in 2020. The authors sampled 254 private and public wells in seven aquifer networks across the Eastern United States for 24 PFAS and various other contaminants. Fourteen of the 24 targeted PFAS species were detected at least once in groundwater samples; at least one PFAS species was detected in more than half of the samples (54%; n=254) and more than two PFAS species were detected in 47% of samples (McMahon et al., 2020). In terms of drinking water well sampled in this study, PFAS were detected in 60% of public water supply wells and 20% of domestic wells. Further, the authors found substantial differences in the frequency of PFAS detections and total (Σ) concentrations of PFAS between the various study well networks with detection frequencies ranging from 3.7 to 92.9% (McMahon et al., 2020). Though compelling in defining source water contamination patterns, it is important to note that this study collected groundwater well supply samples, not samples from the point of use, and so may not be representative of consumer exposure, as PFAS levels may be impacted by treatment processes and distribution/premise plumbing.

A small number of field-scale studies have examined exposure to PFAS in private well-dependent homes at the POU in the US. Mulhern et al. (2021) quantified the effectiveness of a commercially available POU, single stage, activated carbon block (ACB) filter in removing both long and short chain PFAS from 18 homes in North Carolina. All wells and all samples contained at least one detectable PFAS species prior to treatment; the majority (83%) of pre-

treatment influent samples had at least one PFAS above the Minimum Reporting Level (MRL), ranging from 0.5-6.2 ppt (Mulhern et al., 2021). Although this study focused on quantifying the effectiveness of POU carbon activated filters in removing both short chain and legacy PFAS, it is important to note that private well systems commonly do not employ treatment devices (Smith et al., 2014).

Work by Babayev et al (2022) in southeast Alaska demonstrated that drinking water wells located in very rural or remote areas are not immune to PFAS contamination. Twelve PFAS species were detected at least once in the 25 private well water samples collected, with total PFAS concentrations as high as 120 ppt. It should be noted that while the community targeted by this study was quite rural, there was a nearby supply airport.

2.4.2 PFAS detection and quantification in Virginia drinking water systems

At the time of this study, Virginia (VA) remains reliant on federal (US EPA) PFAS exposure guidelines. An initial PFAS screening of municipal water supplies was mandated by the Virginia State Legislature via the passage of HB586 in 2020 (HB586, 2020). This voluntary initial survey was completed by the Virginia Department of Health Office of Drinking Water (VDH) and the PFAS Work Group in 2021. Targeted municipal treatment plants represented the largest systems in the state and/or systems reliant on source waters impacted by identified likely sources of PFAS contamination; 45 of 50 invited systems participated. The initial survey reported that at least one PFAS species was detected in 24% (15/63) of samples collected from 45 municipal drinking water systems across the state, though all samples were below the US EPA's lifetime health advisory level of 70 ppt (VDH, 2021). However, it should be noted that this effort was completed prior to the US EPA reducing the previous Health Advisory of PFOS and PFOA from a combined 70 ppt to 0.004 ppt for PFOA and 0.02 ppt for PFOS in June 2022 (US EPA, 2022), and PFOA and PFOS were detected at maximum values of 5.5 ppt and 7.1 ppt, respectively. In addition, this study analyzed samples according to EPA Method 537.1, which excludes PFBA (US EPA, 2022).

Although this effort did offer some insight into potential human exposures via municipal drinking water, it did not record PFAS levels at the point-of-use or in private drinking water systems. Roughly one-fifth (22%) of Virginia's residents (1.6 million people) are solely reliant on private wells as their primary source of drinking water (). Virginia's Private Well Regulations

(12VAC5-630; in place since 1992) do require testing of newly constructed wells for coliform bacteria but do not require additional sample collection or testing of SDWA-regulated or emerging contaminants (State of Virginia, 1993). Previous examinations of well water contamination in Virginia echo national observations, with indicator bacteria and elevated lead levels regularly observed in samples submitted for analysis to state Cooperative Extension programs (Allevi et al., 2013; Pieper et al., 2015). Testing for emerging contaminants such as PFAS is both expensive and difficult to access for the average homeowner: a single sample costs over \$300 to analyze in a private laboratory and less than five labs in the state offer commercial PFAS testing (VDH, 2021). Because private laboratory issued PFAS results are not publicly available, there is currently no Virginia-specific data quantifying PFAS concentrations at the point of use.

3. Methods

3.1 Site Selection

The study recruited households from Roanoke and Floyd counties as they were within easy driving distance from central Virginia Tech campus, had a history of significant participation in Cooperative Extension well water programming, comprise similar underlying geologies (fractured bedrock), and represent high and low risk for PFAS contamination, respectively, as determined by the Virginia Department of Health Office of Drinking Water (VDH ODW) during their 2021 Phase 1 PFAS Sample Study. A rating of “high” or “low” for a county’s PFAS risk was determined by a variety of factors including the presence of unlined landfills, military or large commercial airports, industrial users, direct dischargers, and other activities that have a potential to involve PFAS (VDH, 2021) (Figure 1). It should be noted that this study did not list the number or physical location of landfills, airports, or industry users in each county. However, according to the VDH study, Roanoke, a high-risk county, contains at least one large commercial or military airport, unlined landfill, and medium sized industry PFAS user, while Floyd contains at least one unlined landfill but no other known potential concentrated sources (VDH, 2021).

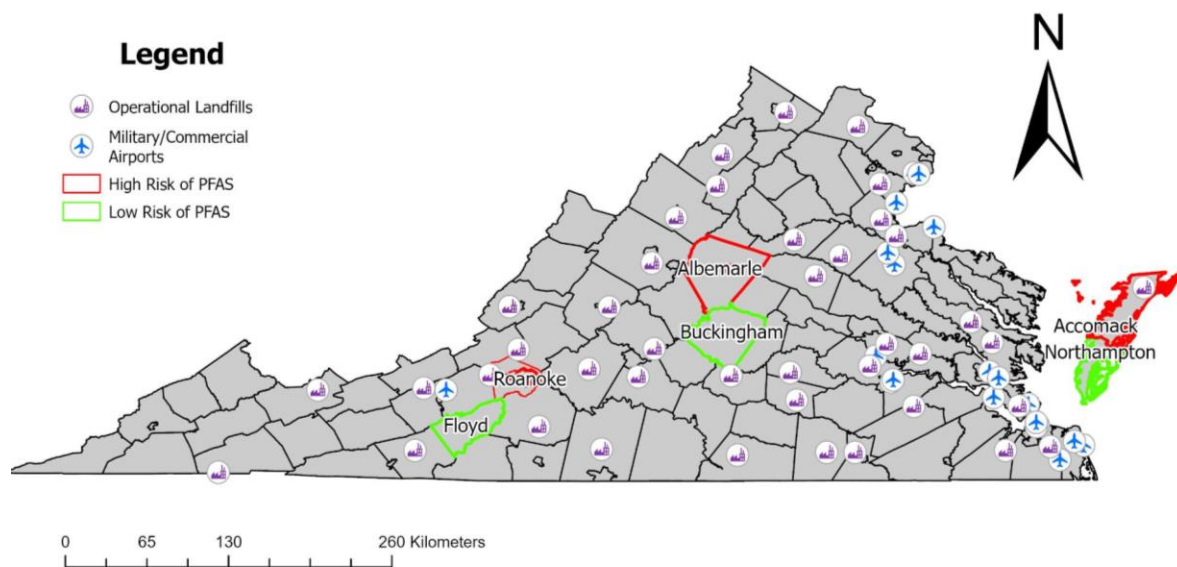


Figure 1. Map of Virginia and its counties with operational, unlined landfills, military, and commercial airports displayed with high and low risk counties highlighted as they were denoted by the Virginia Department of Health. Highlighted counties in the state are the location of current and future PFAS sampling efforts.

3.2 Participant Selection and Testing

3.2.1 Participant Recruitment

Participants for this study were recruited from a list of past participants of the Virginia Household Water Quality Program (VAHWQP) (VAHWQP, 2022). VAHWQP is a Cooperative Extension effort that provides low cost water quality analysis and private water system education throughout the Commonwealth of Virginia (Benham et al., 2016), and has served as a model program for similar Extension-based well water outreach programs throughout the nation (Dameris et al., 2020) Emails inviting participation were sent to all past participants in the last three years who cited their location as within the boundaries of Floyd and Roanoke counties.

Participants were offered PFAS testing as well as a free standard VAHWQP analysis (i.e. for lead, bacteria, etc.) as an incentive for participation. All project recruitment and procedures were approved and overseen by the Virginia Tech Institutional Review Board (IRB #21-492). From the group of responding homeowners, 10 households were then selected in each county for participation based on participant availability during set sampling times.

3.2.2 Point of Use (POU) and Outside Tap Sampling

In order to examine potential bias introduced by homeowner sampling, tap water from each of the twenty homes was sampled twice: once by experts (i.e. graduate students, Extension agents, professors) following the Michigan Department of Environmental Quality PFAS General Sampling Guidance (e.g. specifying clothing, personal care prior to sampling, etc.) (Michigan DEQ, 2021), and once by homeowners following simplified directions and wearing nitrile gloves (Figure 2, Appendix D). All in-home sampling was completed between June and August 2022.

Virginia Household Water Quality Program -- SAMPLING INSTRUCTIONS

- **READ INSTRUCTIONS COMPLETELY BEFORE COLLECTING YOUR SAMPLES.**
 - **Collect your samples ONLY on the collection day you have been assigned.**
 - **Do not remove caps from sample bottles until you are ready to take each sample. Fill each of the 6 sample bottles and replace each cap one at a time as described below.**
 - **Do not write on the sample bottles.**
 - **Store samples in refrigerator or on ice following collection.**
1. In order to test for metals (lead and copper) that might be in your plumbing system, the water will need to remain in the pipes for at least 6-8 hours. **The night before your scheduled collection day, do not use any water after 10 pm.**
 2. Collect the samples first thing in the morning, **before any water has been used anywhere in the house**. Choose a cold-water faucet in the kitchen or bathroom. Be sure that the sink is clean, and that all dishes and other items are removed in order to minimize splashing. If you drink your water, and want to know about the quality of the water you drink, collect from the faucet where you obtain this water. Do not remove the aerator on the faucet.
 3. Put on the blue nitrile gloves from your sampling kit first. Because there are common substances that can interfere with our PFAS analysis **it is important to wear these gloves during the entire sample collection process**. *Because you are using gloves, you will not need to wash your handles prior to collecting water for the bacteria analysis as you have done during regular VAHWQP clinics.*



4. Find the bottle labeled FIRST (with an "X" on the cap) and carefully remove the cap, taking care not to touch the inside of the cap or sample bottle. Do not remove the cap from more than one bottle at a time. Do not put the cap down while collecting the sample.
5. Holding the "FIRST" bottle under the faucet, turn the water on, opening the handle as far as you normally would, and fill the bottle completely. Leave as little air as possible in the bottle, without letting the water overflow. Replace cap when full.

6. Remove the large "PFAS-1" bottle from the double bags. **Do not remove the label placed between the bags.** The special bagging strategy protects the sample from inks or other printing chemicals that may interfere with our analysis while maintaining sample identification. Fill this bottle immediately after the "FIRST" bottle and place back in the inner plastic bag.



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Figure 2. Front page of the instruction sheet included in all homeowner sampling kits (See Appendix D for remaining pages)

Experts traveled to each participant's home in Floyd and Roanoke counties at a pre-arranged time and date. In addition to PFAS, experts collected samples to be analyzed for pH, conductivity, and fecal indicator bacteria (total coliform and *E. coli*) at the indoor point of use (POU) as well as an outside tap (Figure 3). Samples were first collected inside the home at the tap identified as the primary source of drinking water for the household (i.e. kitchen faucet). Conductivity and pH samples were collected in a single prepared 250 mL acid-washed bottle. A single 100 mL sterile bottle was used to collect a sample for bacteriological analysis. Experts then collected a PFAS sample in a 1L high density polyethylene (HDPE) bottle that was washed, prepared, and stored according to Michigan DEQ guidelines (Michigan DEQ, 2021). The same series of three samples were collected by experts outside of the home at a well spicket or faucet depending on the configuration of the homeowner's well system. Samples for PFAS analysis were immediately returned to a double bag and put on ice in a cooler separate from other bottles following collection.

Following expert sample collection, homeowners were provided a home sampling kit that included six bottles for sample collection and sampling instructions (Figure 2, Fig. 4, Fig. 5, Appendix B-D). Residents were instructed to collect their tap water samples from the same inside POU sampled by experts during the initial home visit. Per EPA home sampling guidelines typically used in VAHWQP programming (EPA 815-F-18-022, US EPA, 2018), homeowners were instructed to leave their water stagnant in their pipes overnight in order to obtain a first-draw sample, flush the pipes for 30 s, and collect five additional samples.

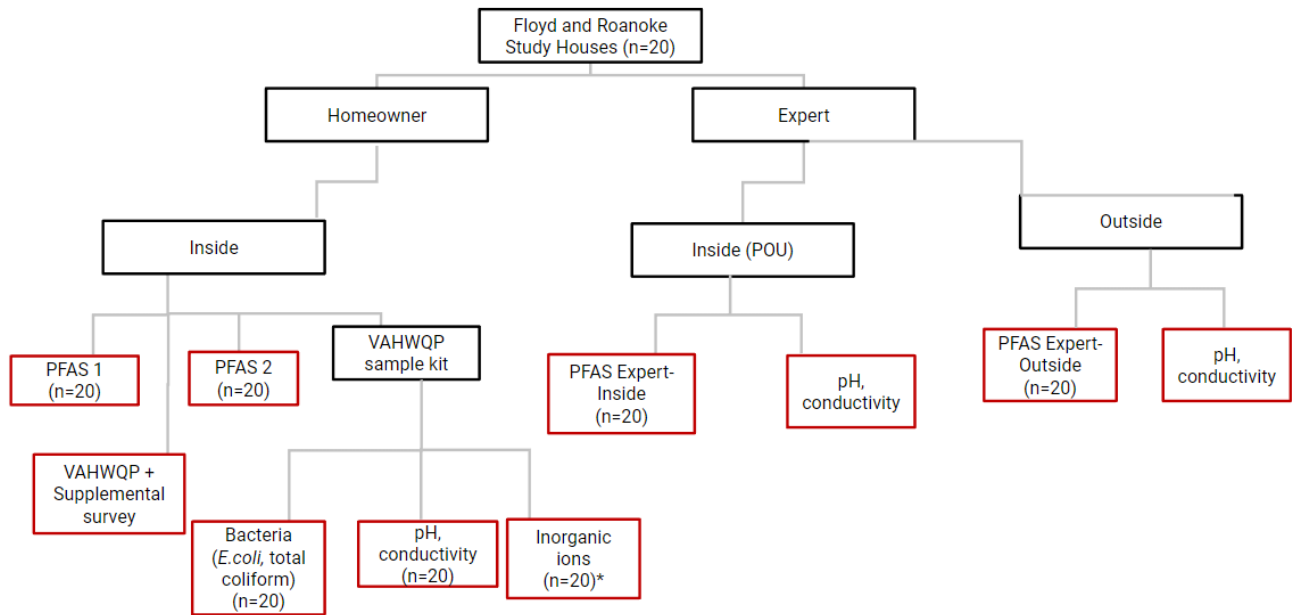


Figure 3. Flowchart of samples collected by homeowners and experts at each study home. (*A first draw and flush sample were collected for inorganic ion analysis so for some analytes, (n=40))

A first draw sample was collected in an acid-washed 250 mL bottle for metals and elemental analysis. Homeowners were then instructed to fill the first PFAS bottle, re-cap, immediately return to double bags. After filling the first two bottles, water was allowed to run for at least a minute to flush out the pipes before filling an additional three bottles for metals, nutrient, and bacterial analysis. Finally, a large 1L HDPE bottle was used to collect the final PFAS sample which was re-capped and immediately returned to double bags. All homeowners were then instructed to place all samples into the refrigerator. Samples were then returned to researchers at a drop-off point within a few hours of collection where PFAS bottles and other analyte bottles were separated and put on ice to be transported back to Virginia Tech for processing. Homeowner sampling all occurred on a single pre-arranged day for each county in keeping with typical VAHWQP sample clinic procedures (Benham et al., 2016).

3.2.3 Participant Surveys

Residents were also provided with two household surveys to complete and return with their water samples (Figure 4, Fig. 5, Appendix B, Appendix C). The VAHWQP survey (Figure 4, Appendix B) consisted of 18 simple multiple choice and short answer questions addressing

perceived water quality, water use, household plumbing characteristics, and alternate drinking water sources and is standard for the VAHWQP program. The PFAS survey (Figure 5, Appendix C) contained 24 multiple choice and short answer questions specifically interested in gathering information pertaining to a household's water treatment system, sociodemographic factors, product use, and prior PFAS awareness (IRB #21-492). Responses were coded in Microsoft Excel and matched to home water quality results.

Virginia Household Water Quality Program - Questionnaire

Biological Systems Engineering Department, Water Quality Laboratory
HABB1 Building – 1230 Washington St SW, Blacksburg, VA 24061
Phone: 540-231-9058 | Email: wellwater@vt.edu | Web: www.wellwater.bse.vt.edu

Sample Number
(LAB USE ONLY):

SAMPLE INFORMATION: To ensure delivery, please print clearly & complete both sides of form.

County (where collected): _____ Date Collected: ____/____/____

Name: _____

Email: _____ Phone: (____) _____
Email must be legible Area code

Mailing Address:

Street address City State Zip code
Sample Location Address (if different from mailing address):

Street address City State Zip code

Sample Point-of-Use/Information for Report (optional): _____
(examples: kitchen tap, outdoor spigot, bathroom, before treatment, after treatment)

BEFORE COLLECTING YOUR SAMPLES:

- Answer the questions below. This information helps us interpret your test results.
- Keep this document DRY.
- Note your sample number for your records.
- Read and follow the included sample collection instructions CAREFULLY.
- Water samples must be collected **ONLY** on the morning of the assigned date. Make sure to bring this questionnaire with your bottles to the drop off location. Contact your Extension office or the Virginia Tech BSE Water Quality Lab at 540-231-9058 with questions.

Water Source Info:

1. What household water supply source was drawn for sample? Choose one:
 Well Spring Cistern Municipal or Public Water Source Other: _____
If well is checked above: (a) Is it a: Drilled Well Dug or Bored Well don't know
(b) What is the well's depth in feet? _____ feet don't know
(c) What year was the well constructed? _____ don't know
2. What water treatment devices are currently installed and functioning properly? Choose all that apply:
 None Don't Know/Not Sure Ultraviolet (UV) Light
 Water Softener (Conditioner) Sediment Filter Reverse Osmosis
 Iron Removal Activated Charcoal Filter Chlorination System
 Acid Neutralizer Other, Please specify: _____
3. How often do you have your water tested? Choose one:
 Never before Once or twice before When I think there is a problem
 Every 5 years Every other year Every year
4. What pipe material(s) is/are used in your house for plumbing?
 Copper Lead Galvanized steel Plastic (PVC, PEX, etc.) Don't Know
 Other → Please specify: _____

Figure 4. First page of the standard VAHWQP survey administered to participants (See Appendix B for remaining pages)

Virginia Household Water Quality Program

Supplementary Questionnaire
HABB1 Building- 1230 Washington St SW, Blacksburg, VA 24061
Phone: 540-231-9058 | Email: wellwater@vt.edu | Web: www.wellwater.bse.vt.edu

Sample
Number
(LAB USE
ONLY)

Principal Investigator: Dr. Leigh-Anne Krometis
Title of Study: Characterizing prevalence and risk factors of PFAS in rural private water
Sponsor: United States Geological Survey

You are invited to participate in a research study. This form includes information about the study and contact information if you have any questions.

WHAT SHOULD I KNOW?

If you decide to participate in this study, you will complete a short survey about your perceptions of your water quality and water use. The study should take approximately 10 minutes of your time. We do not anticipate any risks from completing this study.

You can choose whether to be in this study or not. If you volunteer to be in this study, you may withdraw at any time without consequences of any kind. You may also refuse to answer any questions you don't want to answer and remain in the study. The investigator may withdraw you from this research if circumstances arise which warrant doing so.

CONFIDENTIALITY

Any data collected during this research study will be kept *confidential* by the researchers. Your answers will be linked back to your water quality data by your lab code, but will not be linked to your name or address. Please *do not include your name* or other identifying information in your responses that can identify you so we can be sure to keep your answers as confidential as possible.

WHO CAN I TALK TO?

If you have any questions or concerns about the research, please feel free to contact Dr. Leigh-Anne Krometis at 540-231-4372 or krometis@vt.edu. You are not waiving any legal claims, rights or remedies because of your participation in this research study. If you have questions regarding your rights as a research participant, contact the Virginia Tech HRPP office at 540-231-3732 ([HYPERLINK "mailto:irb@vt.edu" irb@vt.edu](mailto:irb@vt.edu)).

WATER CHARACTERISTICS: Please answer the following questions based on how your water is now.

1. Does your water have an unpleasant taste? yes no
 If YES, how would you describe the taste? (*check all that apply*)
 bitter sulfur salty metallic oily soapy other: _____
2. Does your water have an unpleasant odor? yes no
 If YES, how would you describe the odor? (*check all that apply*)
 rotten egg/sulfur kerosene or gas musty chemical other: _____
3. Does your water have an unnatural color or appearance? yes no
 If YES, how would you describe the color or appearance? (*check all that apply*)
 muddy milky black/gray tint yellow tint oily film other: _____

Figure 5. First page of the supplemental PFAS survey administered to participants (See Appendix C for remaining pages)

3.3 Analytical Methods

3.3.1 PFAS Analysis

PFAS analysis (Appendix A) of water samples collected by homeowners and experts both inside and outside the home were analyzed on an Agilent LCMS 6490 Triple Quad System according to US EPA Standard Methods 533 and 537.1 via direct injection (US EPA, 2022). Samples were analyzed using direct injection rather than solid phase extraction in order to quantify the actual PFAS exposure at the tap. As previously discussed, current US EPA health advisories for PFOA and PFOS are below the current detection and quantification limits for typical analytical equipment (US EPA, 2022; Virginia Department of Health, 2022; Teymoorian et al., 2023). As a result of quantification limits on the LCMS machine at the time, coupled with analytical standard availability, the detection limit for PFOA, PFOS, and PFBS in this instance is 25 ppt; the detection limit for all other analytes is 5 ppt. Detections below the lowest analytical standard were extrapolated using the established calibration curve. These extrapolated values are below the quantification limit (BQL); however, as current regulatory levels are below BQL, and in order to increase the amount of data available for comparison to established health advisories, levels BQL were used in subsequent statistical analyses. Quantification limits are noted, and extrapolated BQL values are denoted via asterisks in all Tables and Figures.

3.3.2 Standard Water Analyses

In addition to PFAS, samples were analyzed within 12 hours of collection for total coliform bacteria and *E. coli* via the Colilert defined substrate method (IDEXX, Westbrook, MN; Standard Method 9223B) (Table 2, Appendix G, H). Concentrations of metallic cations (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Fe, Mn, Co, Ni, Cu, Zn, As, Se, Sr, Mo, Ag, Cd, Sn, Ba, Pb, U) in tap water samples collected at the POU by homeowners were determined via the ICP-IMS process as described in Standard Methods 3030D and 3125B (APHA/AWWA/WED, 1998). Standard Methods 4500-NH was used to assess NO₃ concentrations in the water samples and Standard Method 300.0 was used to assess concentrations of F (APHA/AWWA/WEF, 1998). Both pH and conductivity were determined in the laboratory via an Oakton benchtop pH/conductivity/TDS meter (Cole Parmer, Vernon Hills, IL, USA).

3.3.3 Statistical Analysis

All statistical testing was conducted in RStudio version 12.0 +353 (RStudio, Boston, MA). Normality was tested using the “qqnorm” command in the “ggplot” data package. After confirming a non-normal data distribution, a Wilcoxon Signed Rank Test was run on the constituents to determine difference between total PFAS concentrations in homeowner and expert collected samples with a significance defined at an alpha value of 0.05 (Table 3). A Spearman’s correlation matrix was prepared to identify potential statistical relationships between maximum values of the study’s most commonly detected PFAAs and inorganic ions of human health concern (Figure 9, Appendix S).

3.3.4 Homeowner Results Letters

In keeping with the mission of the VAHWQP and Cooperative Extension, all participating homeowners received copies of their water quality results via mail or email, depending on preference. Standard water quality measures (coliform, lead) were compared to existing EPA regulatory guidelines for municipal systems per standard VAHWQP communication (Benham et al., 2016). Because both the analytical and regulatory landscape for PFAS is rapidly evolving, homeowners received separate letters outlining their PFAS results (Figure 6, Appendix E, F).



Biological Systems Engineering
155 Ag Quad Lane
Seitz Hall, Room 308
Blacksburg, Virginia 24061
P: (540) 231-4372 F: (540) 231-3199
krometis@vt.edu
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December 5, 2022

Dear [REDACTED]:

Thank you for participating in our United States Geological Survey (USGS) funded pilot study investigating the presence of per and poly-fluoroalkyl substances (PFAS) in private household wells (Virginia Tech study IRB #21-492). This is the *first known* effort in Virginia to systematically analyze private drinking water samples for PFAS. Your participation is essential in helping us understand best practices for collecting samples before we begin larger scale sampling efforts in your county.

*The results from your water samples are attached. All samples were directly injected onto a Ultraperformance Liquid Chromatography/Tandem Mass Spectrometry (LC/MS/MS) and analyzed for a total of 30 PFAS compounds using EPA methods 533 and 537.1. Please note that there are many different types of PFAS chemicals, and there is limited understanding of the risks that most of these pose to human health. At present, the United States Environmental Protection Agency (USEPA) has issued *final health advisories* for two specific PFAS chemicals (GenX, PFBS) and *interim health advisories* for two additional specific PFAS chemicals (PFOA, PFOS). You can read more about EPA's process for investigating PFAS impacts and potentially establishing associated regulatory limits under the Safe Drinking Water Act here: [Questions and Answers: Drinking Water Health Advisories for PFOA, PFOS, GenX Chemicals and PFBS | US EPA](#). The remaining types of PFAS we examined in your sample are recommended for monitoring, but are *not* currently associated with health advisory limits in municipal systems. Because PFAS chemical types are often proprietary, the primary sources of many of these chemicals is unknown. Your participation in our study is allowing researchers at Virginia Tech and USGS to begin to identify and understand potential relationships between typical household products, plumbing systems and PFAS in drinking water.*

Over the course of this study we collected three samples from your household tap (i.e. point of use): Kathleen Hohweiler (our graduate student) collected one sample on her initial visit, and then you collected two samples in large bottles that you brought to the Extension office. We did not observe any statistically significant difference between graduate student- and homeowner-collected samples. To be as clear as possible, we provide the maximum measured level from these three samples for comparison to the health advisory limits.

The regulatory landscape for PFAS is changing regularly for municipal water supplies and is challenging at present. It is very likely that some types of PFAS will be regulated in municipal water supplies in the near future through the Safe Drinking Water Act

VIRGINIA POLYTECHNIC INSTITUTE AND STATE UNIVERSITY
An equal opportunity, affirmative action institution

Figure 6. First page of the PFAS results letters returned to participants (See Appendix E, F for remaining pages)

In addition to household specific results, these letters included final and interim health advisories for GenX, PFBS, PFOA, and PFOS as well as additional links to US EPA resources for questions regarding treatment options. The compounds name, abbreviation, health advisory (if applicable), and the maximum detected PFAS value across the three indoor POU samples collected (two by the homeowner and one by the expert) was provided for each species. Maximum values across the three samples were chosen for reporting as it represents the highest level of potential exposure at the tap and is so most conservative in protecting public health. If the detected value was above the quantification limit, PFAS results were reported in parts per trillion (ppt). Samples that returned a value between 0 ppt and 5 ppt were reported as “BQL” to indicate there was a detection but without reporting a value with lower confidence. PFAS species that were not detected by the instrument were reported as non-detect, “ND”.

4. Results and Discussion

4.1 Household System Design and Water Perception

A total of 19 participants completed the standard VAHWQP survey (Figure 4, Appendix B) describing basic system characteristics and perception of in-home water quality. All confirmed they were reliant on private wells or springs. The average well sampled in this study was constructed approximately 24 years ago and is 325 feet deep. Three households reported their private system source as a spring. Almost half (47%) of respondents reported living within a mile of a farm animal operation. Less common nearby contributors reported by homeowners included landfills (5%), fruit orchards (5%), and manufacturing operations (5%). Most household systems were built with plastic piping (79%) though almost half of homes with plastic pipes indicated they also had some copper piping (40%). A majority of homes (65%) reported using some kind of water treatment, with sediment filters (58%) and water softeners (32%) being the most commonly reported. Other reported treatment mechanisms included ultraviolet light devices (16%), acid neutralizers (11%), reverse osmosis (5%), iron removal (5%), and carbon filters (5%).

All but two households (n=17; 89%) reported that they drink their household water. Only one respondent reported someone in the home becoming sick after drinking the water. Six homes reported a perceived water quality issue (i.e. abnormal taste, smell, or appearance), including corrosion (16%), staining (21%), visible particles (21%), or unpleasant smell (15%), taste (20%), and appearance (20%) With respect to unpleasant odor associated with their home drinking

water, “sulfur”, “musty”, and “chemical” were the chief complaints. “Metallic” and “sulfur” were the most commonly cited taste descriptors by respondents. Although only 20% of homeowners reported their tap water having an unnatural appearance or color, half of these homes (50%) indicated that their water had more than one negative visual descriptor with “yellow tint” and “oily film” being the most commonly cited, followed by “muddy” and “black/grey tint”.

4.2 Household Demographics and Potential PFAS Sources

Although all homes answered at least some of the questions in the supplemental PFAS survey (Figure 5, Appendix C), only 18 of the 20 homes responded to all of the survey questions. All participating households (100%) reported owning their homes. On average, each household included 2.5 individuals (i.e. 55 reported household members across 20 homes), with 86% of them self-reporting as white. More than half of the households in this study (55%) included at least one member that has earned a post-college degree (i.e. MS, PhD, etc) and almost half (45%) report an annual household income over \$98,000. These demographics are in keeping with past examinations of participants in this program (Smith et al. 2014).

As was reported in the VAHWQP survey, 89% of respondents drink their system water. Three homes used a point of use filter (e.g. a faucet mounted, under the sink reverse osmosis, or refrigerator filter). Roughly one-third (35%) of households reported purchasing bottled water for drinking, with 20% reporting that they purchase bottled water daily. All survey respondents (100%) indicated that they use their system’s water for at least one household task with cleaning (100%), cooking (95%), bathing (95%), and brushing teeth (95%) being the most frequently reported.

Homeowners were asked to indicate if they lived within half a mile of a list of previously identified industries and facilities that commonly produce, use, or store products that are known to contain PFAS such as firefighting facilities, areas with land applied biosolids, airports, and military installations. Only one household indicated that they lived within half a mile from a potential source (carwash); however, almost half (47%) of homeowners reported living near an animal farm facility.

Homeowners estimated their households’ usage of a variety of products that are suspected to contain significant amounts of PFAS. The most common “daily” usage products

identified were nonstick cookware and cosmetics (both 37%). Households also reported “occasional” use of waterproofed clothing (i.e. GoreTex) (79%), aluminum foil (63%), sunscreen (58%), microwave popcorn (47%), and microwave meals (26%). Cleaning and laundry products such as stain treatments for carpet or upholstery and dryer sheets were used the least frequently, with 74% and 58% of respondents indicating these products are “never” used in their home.

PFAS, particularly legacy contaminants such as PFOA and PFOS, have gained increasing media and public attention as a result of the US EPA’s aggressive interim health advisories and a well-publicized illegal discharge of PFOA by DuPont in the mid-Ohio Valley (Herrick et al., 2017). Even so, less than half of the participating households had heard about PFOA/PFOS in the national news (44%) and only two households had heard about PFAS in state or local news (11%). Three participants included notes at the end of their survey stating that they did not know what PFOS/PFOA were, emphasizing a need for clear communication. Interestingly, only a few participants (39%) reported having seen products advertised as PFOA/PFOS free, but the majority (89%) indicated that the designation of a product as “PFOA/PFOS free” would influence their decision to buy it.

4.3 Bacteriological Contaminants

No samples (n=20) tested positive for *E. coli*, and less than half of samples in both counties tested positive for total coliform with an overall average coliform value of 560.3 MPN/100mL in positive samples (Table 2). Slightly more samples from homes in Floyd (40%) as compared to those from homes in Roanoke (20%) County were coliform positive. The associated US EPA SDWA standard for municipal systems is zero, i.e. coliform absent.

Bacteriological Contaminants		Floyd County (n=10)				Roanoke County (n=10)			
		Max	Average	# in violation	% in violation	Max	Average	# in violation	% in violation
M C L	Total Coliform (0 MPN/100 mL)	1709	834	4/10	40%	21.7	13.5	2/10	20%
	<i>E. coli</i> (0 MPN/100 mL)	ND	ND	ND	0%	ND	ND	ND	0%

Table 2. Results from bacteriological analysis from study homes in both counties.

However, considering the likely interactions between soil and groundwater in a private system as well as a lack of disinfecting treatment in more homes, the presence of coliforms is unsurprising. Interestingly, one of the six homes that tested positive for total coliform employed a treatment system rated to remove bacteriological contaminants (i.e. chlorinator, UV light). Previous literature consistently indicates that bacteriological contamination is the most common health-related water quality issue in samples sourced from private systems (Allevi et al., 2013; Patton et al., 2020). The absence of *E. coli*, which would indicate a more serious risk of infectious disease (Paruch & Mæhlum, 2012), is noteworthy, as it is typically recovered from 1 in 10 samples collected by this Extension effort (Allevi et al., 2013; Pieper et al., 2015; Smith et al., 2014). Previous assessment suggests that participation in VAHWQP programming is associated with subsequent improvements in system maintenance (Benham et al., 2016); as participants in this study were all previous VAHWQP participants, they do represent a more informed and potentially more conscientious population.

4.4 Inorganic Ion Contaminants

All 20 households enrolled in this study collected a first draw and a flush tap water sample for US EPA regulated inorganic metal cations and anions as part of their compensated VAHWQP kit. No samples collected from any home in the study exceeded associated health-based (MCL) standard values for municipal waters (Appendix G, Appendix H). Samples from two homes in Floyd and one home in Roanoke County exceeded the Treatment Technique recommendations for Cu, which could indicate an issue with corrosive source water. Although no homes exceeded the MCL for Pb (> 15 ppb), it should be noted that the US EPA has set the Maximum Contaminant Level Goal for lead at 0 ppb. More than 80% (82.5%) of in-home water samples contained detectable lead in either the first draw or flush sample (n=40), with a maximum observed value of 10.2 ppb. Al, Fe, and Mn were occasionally detected in samples at values exceeding recommended taste and aesthetic levels (SMCL) in homes in both counties. Interestingly, 70% of homes in Roanoke and 20% of homes in Floyd County yielded samples that exceeded the SMCL for Na. Increased consumption of sodium can be an issue for those on a low sodium diet or with chronic medical problems such as hypertension or high blood pressure (Thompson et al., 2022). Na is a naturally occurring cation often associated with wells drilled in areas with bedrock erosion taking place. Roanoke County in particular is located in the Valley

and Ridge where the bedrock consists of sandstone, limestone, dolomite, and shale, commonly sodium-bearing rocks (VDH, 2022). Further, sodium is frequently used in water softeners, which are commonly employed water treatment devices, to remove high concentrations of iron or to reduce water hardness.

4.5 PFAS incidence in point of use (POU) samples

At least one PFAS species was detected in 76% of indoor POU drinking water samples (n= 60). On average, the Σ PFAS concentration in POU samples across both study counties was 23.5 ppt, though average Σ concentrations in a single sample as high as 160 ppt were recovered (Figure 7, Table 4, Appendix I, J, K). The most commonly detected species associated with current regulatory recommendations at the POU in Roanoke County homes was PFBS, a short chain PFOA replacement compound, which was detected in a majority (80%; 24/30 samples) of samples. However, no samples in either county containing PFBS exceeded the US EPA final health advisory set at 2,000 ppt and the average concentration for both counties was below the detection limit (4.3 ppt in Roanoke and 1.07 ppt in Floyd). Less than half (43%, 13/30 samples) of POU samples collected in Floyd County contained trace concentrations of PFBS, with a maximum concentration below the detection limit (2 ppt). The most commonly detected PFAS in Floyd County samples was the short chain, replacement PFAA, ADONA, which was detected in 63% of samples (19/30 samples), with the average concentration also below the level of detection (1.2 ppt). The detection of short chain PFAAs was common at the POU in both study counties. Short chain PFCAs and PFSAs, though only accounting for 7 of the 31 PFAS analyzed, were responsible for 65% of detections in POU samples (n=30). Long chain PFCAs, PFSAs, FOSA, and other PFAS combined contributed the remaining 35% of total POU detections across both study counties. Only 3 of the 60 POU samples collected included a Σ PFAS value that exceeded the 2016 US EPA lifetime health advisory of 70 ppt. Notably these three detections represent three separate homes meaning three homes each submitted one POU sample with a Σ PFAS \geq 70 ppt and two other samples below this threshold. This reflects potential large variations in PFAS recovery; in the case of two of these homes (#3 and #5), concentrations in all other samples were below the average Σ PFAS observation of 23.5 ppt (Figure 7).

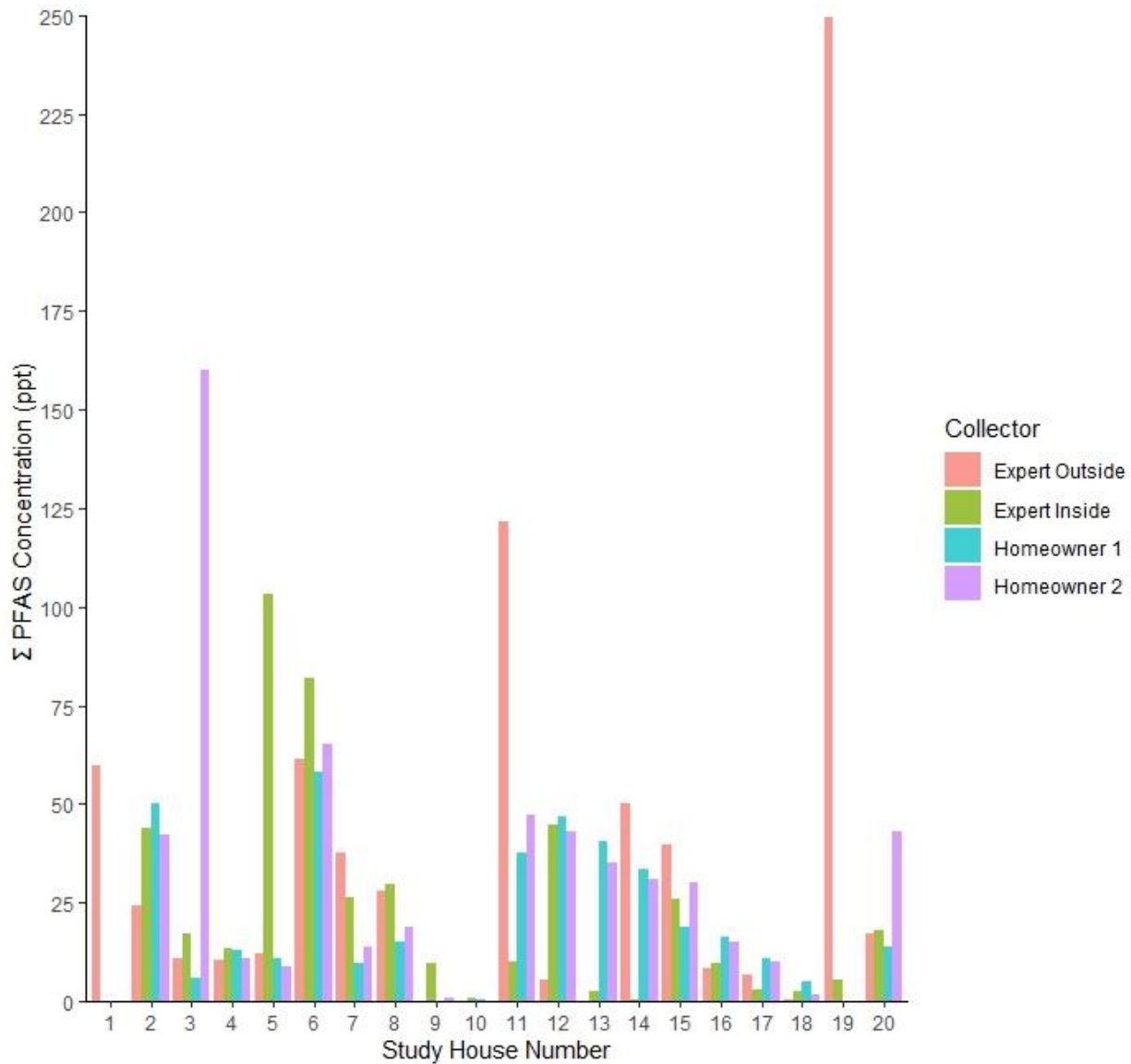


Figure 7. Σ PFAS concentrations per study household in both counties (ppt). Homes #1- #10 are in Roanoke County and homes #11-#20 are in Floyd County (See Appendix O, P for individual county graphs)

Additional commonly detected PFAS in POU samples (n=60) are all short chain replacements or fluorinated chemical degradation products: PFBS (62%), PFBA (48%), NaDONA (42%), and PFHpA (37%). Interestingly the replacement PFAS commonly used in AFFFs, 6:2 FTS, was detected in significant concentrations in several samples from Floyd County homes (average value: 17.2 ppt) though the compound was not detected in any Roanoke County homes. This is unexpected, as Roanoke County includes an airport, while Floyd County does not, however groundwater contamination can extend across county boundaries

It is worth noting that hexafluoropropylene oxide dimer acid, or GenX, a short chain replacement species associated with an EPA final health advisory, was not detected in any samples. Many long chain species of human health concern including PFDoS, PFDoA, PFDA, and PFNA were also not detected in any POU samples (Appendix N). However, the two most well studied, PFOA and PFOS, were detected in 13% and 22% of samples (n=60), respectively. Average concentrations for both PFOA and PFOS (7.4 and 3.1 ppt) were below the lowest analytical standard available at the time of this study (25 ppt) and so these averages represent extrapolated values. However, it is worth noting that current Interim Health Advisories for these compounds are essentially below quantifiable limits (0.004 ppt and 0.02 ppt, respectively), so any detection is practically considered above this recommendation.

4.6 Comparing PFAS incidence in expert and homeowner collected samples

Because various PFAS are commonly detected in a variety of personal care and consumer products, it is very easy for sample contamination to occur during any step of sample preparation, collection, or analysis. It was hypothesized at the beginning of this study that homeowners may not be able to collect their own samples for PFAS analysis in future sampling efforts due to the high risk of sample contamination. In order to test this hypothesis, expert sample collectors in this study followed the rigorous sampling guidelines recommended by the Michigan Department of Environmental Quality (e.g. all cotton clothing; no personal care products for 24 hours prior to sample collection, etc) while homeowners were given written instructions that simply required the use of nitrile gloves and careful handling of PFAS-free sampling bottles (Figure 2, Appendix D). Homeowner and expert samples in both counties were matched using their unique VAHWQP sample kit ID number. Roanoke and Floyd county sample results were also combined in order to increase the number of sample pairs available for analysis. Reported concentration values of analyzed PFAS species in homeowner and expert collected samples were summed to get a total PFAS concentration for each sample. Pairs of expert and homeowner samples where both totals were “ND” were excluded from analysis. Because homeowners collected two POU flush samples (n=40) while experts only collected one inside sample at every home (n=20), the Wilcoxon Test was run twice to separately compare expert samples to both groups of homeowner samples. Results indicate that while total PFAS concentrations between the two homeowners’ collected samples were equivalent, concentrations in both of these samples were statistically different from those collected by experts (Table 3).

However, interestingly, this bias was inconsistent: average sum concentrations of PFAS in expert samples were on average greater than the first homeowner sample collected, and less than the second. Therefore, the source of this bias is unclear, and seems more likely related to the timing of sample collection rather than contamination by the collector. Homeowner samples were collected first thing in the morning so as to include a first draw sample for metals in keeping with standard VAHWQP practice. In contrast, the majority of expert samples were collected mid-day or late afternoon when homeowner and expert availability best aligned. Changes in water quality from private systems over the course of a single day, particularly for metals, is well established (Pieper et al., 2015); however, potential patterns of PFAS detection over time have not been examined. Sampling at different times may reflect differences in water age or origin (e.g. stagnant premise plumbing, pressure tank, source water) and is deserving of further investigation.

4.7. Comparing PFAS incidence in Roanoke and Floyd Counties

It was hypothesized at the beginning of this study that samples collected at the POU in Virginia counties that contained PFAS-producing or legacy sites would be at a higher risk for PFAS contamination (Figure 1). The data was determined to be non-parametric, so a Wilcoxon Rank Sum Test was used to determine the relationship between Σ PFAS concentrations in Roanoke and Floyd Counties (Table 3). Although the average Σ PFAS concentration in Roanoke POU samples was higher (28.9 ppt) than in Floyd POU samples (18.8 ppt), these differences were not statistically significant (Wilcoxon, alpha= 0.05).

Hypothesis	Test Outcome (alpha= 0.05)	Result	Directionality
Ho: Expert and homeowner sample #1 Σ PFAS concentrations are the same	Significantly different	Expert and homeowner sample #1 Σ PFAS concentrations are statistically different	Expert > Homeowner
Ho: Expert and homeowner sample #2 Σ PFAS concentrations are the same	Significantly different	Expert and homeowner sample #2 Σ PFAS concentrations are statistically different	Homeowner > Expert

Ho: Homeowner collected samples #1 & #2 Σ PFAS concentrations are the same	Fail to reject the null	Not enough evidence to conclude the two groups have statistically different total concentrations	NA
Ho: The number of detected PFAS compounds in homeowner samples #1 and #2 are the same	Fail to reject the null	Not enough evidence to conclude the number of detectable PFAS compounds varies between the two samples	NA
Ho: Expert inside and outside Σ PFAS concentrations are the same	Fail to reject the null	Not enough evidence to conclude Σ PFAS concentrations in expert inside and outside samples vary	NA

Table 3. Hypotheses, test outcomes, and results of the various Wilcoxon Ranked Sum Tests used to examine the relationship between homeowner and expert collected samples in terms of Σ PFAS concentrations.

The average number of distinct PFAS species detected in each sample was similar across Floyd and Roanoke Counties at 3.4 and 3.2 species detected, respectively (Table 4). Roanoke County POU samples had slightly less detections of sample mixtures containing ≥ 3 PFAS (66%, n=30) than Floyd county (70%, n=30).

Sample Group	n	PFAS Sum Values			
		Avg. Σ PFAS concentrations (ppt)	Max Σ PFAS concentrations (ppt)	Avg. # of PFAS species detected*	Max # of PFAS species detected*
Floyd County POU	30	18.8	47.4	3.2	6
Roanoke County POU	30	28.9	160	3.4	8
All Study Homes- POU	60	23.5	160	3.3	8
Floyd County - Expert POU	10	12.2	44.8	3.1	6
Roanoke County- Expert POU	10	32.6	103.2	3.2	7
Floyd County- Homeowner Sample #1	10	20.2	46.7	3.1	6
Roanoke County- Homeowner Sample #1	10	18.1	58.1	3.4	6
Floyd County- Homeowner Sample #2	10	23.3	47.4	3.5	6
Roanoke County-Homeowner Sample #2	10	35.6	160	3.5	8
Floyd County- Expert Outside	10	49.9	249.5	3.2	6
Roanoke County- Expert Outside	10	24.4	61.3	3.6	8
All Study Homes- Outside	20	37.2	249.5	3.4	8

Table 4. Σ PFAS concentration and species results.

4.8 Comparing PFAS incidence in paired point of use (POU) and outside tap samples

The detection of PFAS, particularly in mixture, was common in samples collected directly from outside taps in both Floyd and Roanoke counties (Figure 8). Samples collected from outside taps in Floyd County had nearly twice the average Σ PFAS concentration (49.9 ppt) as compared to those collected from homes in Roanoke County (24.4 ppt) (Table 4). These detections may indicate the presence of environmental or geological factors contributing to PFAS presence in Floyd if the outside taps represent pre-treatment and pressure tank water. However, given the heterogeneity of indoor plumbing layouts, these high detections might also represent simple changes across plumbing types, the presence of garden hoses or well caps that may be sources of PFAS. There were several notably large detections of Σ PFAS concentrations in outside tap samples from both Floyd and Roanoke systems (Figure 7). The overall study maximum Σ PFAS detection was from a private system in Floyd county (249.5 ppt). This particular sample had a mixture of ≥ 3 PFAS that comprised of only short chain PFAAs. Further, this mixture contained the maximum detection of PFBS seen across the entire study effort (202.5 ppt).

On average, samples collected from outdoor taps in Roanoke had slightly more PFAS species in mixture per sample (3.6) than those collected from outdoor taps in Floyd (3.2) (Table 4). However, again, there were more notable complex mixtures in Roanoke and Floyd homes. An outside sample from one Roanoke home included detectable levels of 8 PFAS, including a mix of both legacy and replacement species. PFBS was detected in more than half of samples from outside taps (65%, n=20). PFOS was only detected in four samples (two per county) and the average concentration being under the quantification limit for this project (3.95 ppt) and a maximum of only 7.2 ppt. Similarly, PFOA was detected in five outdoor POU samples but at an average level BQL (2.42 ppt; max = 10.1 ppt). There was no significant difference between Σ PFAS concentrations in samples collected by experts at the inside vs outside POU (Wilcoxon Ranked Sum Test, Table 3). However, there were instances of homes that had no detectable PFAS in outside tap samples but had rather complex mixtures in samples collected at the POU. For example, an outside tap sample from a Floyd County home #13 had no detectable PFAS but as many as six PFAS detected and a maximum observed Σ PFAS concentration of 40.5 ppt was recovered from an indoor POU sample collected at the same home (Figure 7).

4.9 Examining the incidence of complex PFAS mixtures

As mentioned previously (Section 2.3.2), only four species of PFAS are associated with official US health advisories, but simultaneous exposures to mixes of different species is of increasing concern (US EPA, 2022). In this study, 66% of analyzed samples (53/80 samples) contained ≥ 3 PFAS species (Table 4). The average number of PFAS in a POU sample was 3.3 while the average in outside samples was 3.4 (Table 4). The maximum number of PFAS detected in mixture at the POU was 8 species in a Roanoke County home. Several PFAAs were frequently detected together in mixture in POU samples (Figure 8). Short chain PFCA, PFBA, and short chain PFSA, PFBS were detected together 21 times out of 60 samples. 21 of the 60 POU samples collected contained PFOA or PFOS but PFOA and PFOS were detected together in samples 7 times. Similarly, legacy PFAAs and their designated replacement short chains were also detected together in mixture in the same POU samples. PFOS and its short chain replacement PFBS, were detected together in 10 of the 60 total POU analyzed samples. PFOA also co-occurred with other various PFCAs including PFHxA and PFBA, although seemingly less frequently. Unlike PFOS + PFBS, PFOA + PFBS were detected together in only one sample and PFOA + PFHxA were detected together in 4 samples.

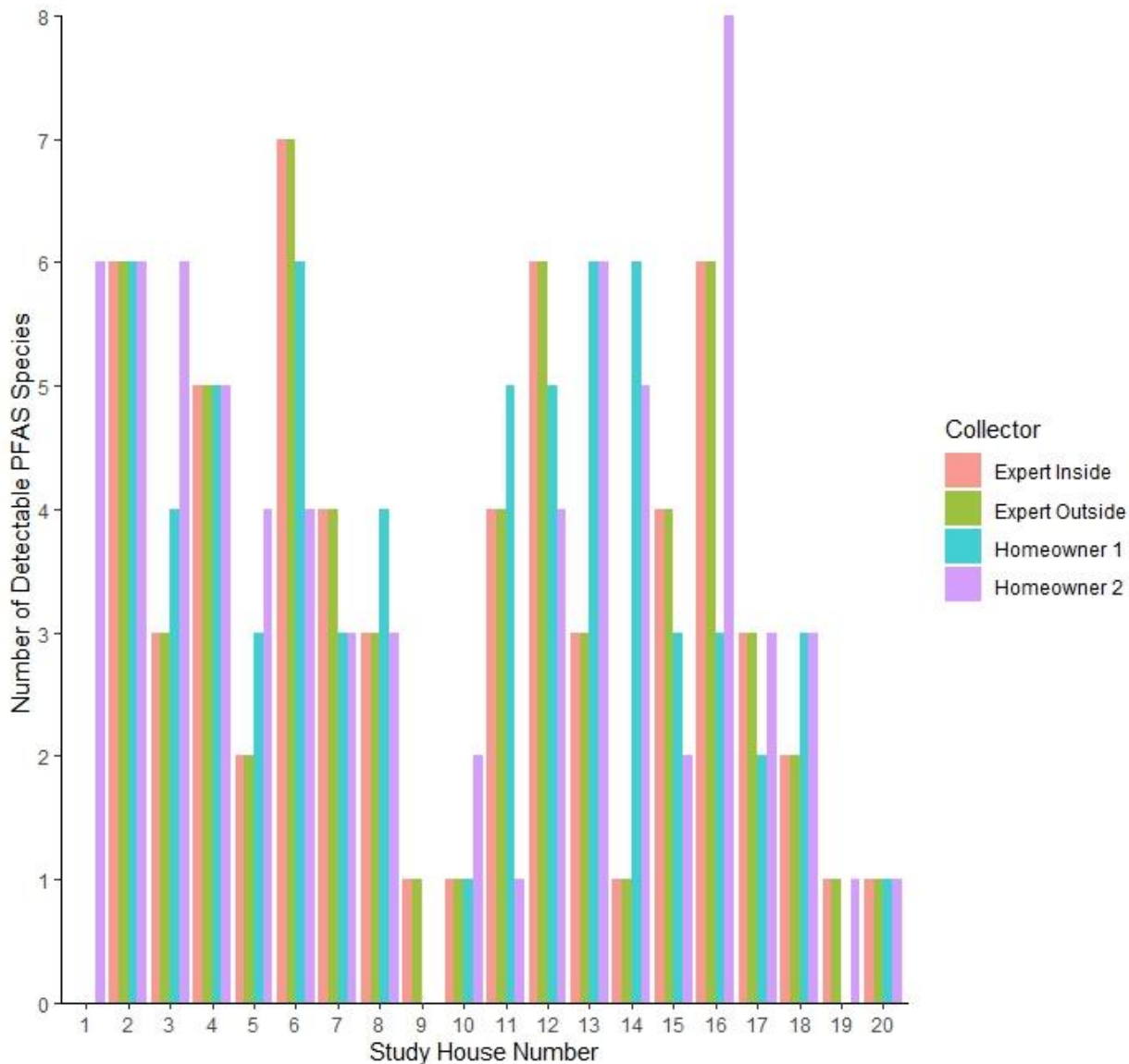


Figure 8. Number of PFAS species detected per sample per study household in both study counties. (Homes #1-#10 are in Roanoke County and homes #11- #20 are in Floyd County; See Appendix Q, R for individual county results)

A Spearman’s Correlation Matrix visualizing the most commonly detected PFAS in samples (occurred in > 20% of samples), as well as potential co-occurrence with additional contaminants of interest to human health, was completed (Figure 9, Appendix S). The strongest positive relationships were between PFOS + PFHpA which were detected in 8 of the same samples (0.73). As previously mentioned, only 13 samples had detectable PFOS, meaning 62% of detects also had a PFHpA detect. This is interesting considering PFOS and PFHpA are not in the same family of PFAAs and PFOS is considered to be a legacy contaminant while PFHpA is

still in production. Other strong significant relationships between PFAS include PFHxA + PFHpA (0.68) which also occurred together in 15 of the 22 samples (68%) with PFHpA detects. Magnesium and PFAA species, ADONA, had the strongest positive relationship between a PFAS and metal (0.67). On the other hand, however, fellow short chain, PFHxS, had a relatively strong negative relationship with magnesium (-0.55). Furthermore, PFOA and lead were also somewhat negatively correlated (-0.51), possibly representing the influence of the drinking water system treatment. PFBA and PFOS had a strong negative relationship (-0.57) which is supported by the notion that they were only detected together in mixture once. This finding is further supported by the fact that the species PFBA was specifically created in order to replace PFOS in typical industry products (ITRC, 2020).

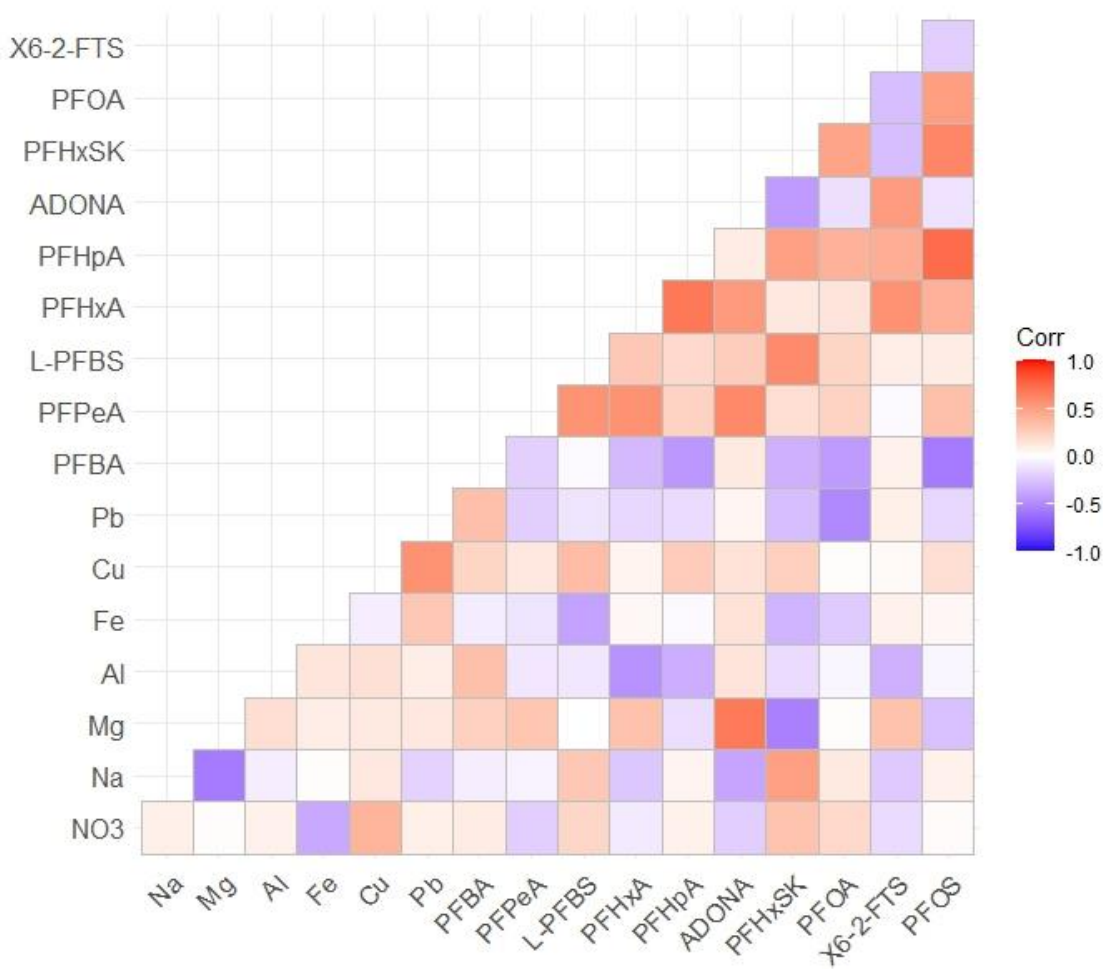


Figure 9. Spearman's correlation coefficient matrix values comparing maximum concentration values of commonly detected PFAS and inorganic ions across all POU samples (n=60)

5.0 Conclusions

The goal of this project was to study the incidence of per and polyfluoroalkyl substances in private drinking water supplies in two Southwest Virginia counties, Floyd and Roanoke, and to determine if it was possible to employ citizen science initiatives for PFAS sample collection in subsequent, larger sampling efforts. Overall, 76% of POU samples (n=60) contained at least one detectable PFAS, with an average Σ PFAS concentration of 23.5 ppt, indicating that the recovery of legacy and replacement species in samples from private drinking water systems is fairly common. The rate of detection is three times that reported in the 2021 VDH study of Virginia municipal drinking water systems (24% of samples positive; i.e. 15/63). However, it is worth noting that in keeping with regulatory needs, analytical standards are advancing rapidly and reducing detection limits (Teymoorian et al., 2023). The VDH study did not list the value of their practical quantitation level (PQL) and their samples were analyzed after undergoing solid phase extraction which would concentrate the sample and remove all suspended solids that may have PFAS absorbed to the minerals suspended in solution. Further, these researchers were preparing their effort under the guise of the 2016 US EPA health advisory for PFAS, Σ PFAS should be < 70 ppt. This study relied on direct injection, which represents a slight variation on US EPA methods 533 and 537.1, in order to better represent the actual composition of drinking water being consumed by homeowners. The role that dissolved minerals play in the sorption of PFAS in untreated groundwater used for drinking has not been fully explored (Hellsing et al., 2016).

Almost half of the PFAS species targeted by LC/MS analysis (15/31) were detected at least once from homeowners' private drinking water systems. Two thirds of samples (53/80) contained ≥ 3 PFAS in mixture (Figure 9, Appendix S). Relatively little is known about the scope of PFAS mixture occurrences in drinking water. However, laboratory studies have demonstrated that complex mixtures of specific PFAS, including PFHxS, GenX, PFNA, and PFBS, may have additive toxic effects. An examination of patterns of PFAS detection in groundwater wells along the east coast of the US noted that mixtures occur frequently; ≥ 3 PFAS occurred in > 80% of mixtures but the dominant PFAS in mixtures varied between sampled well networks (McMahon et al., 2020). The present study echoes these findings as the average indoor POU sample contained 3.3 species and the average sample sourced from an outdoor tap contained 3.4 species (Table 4).

Although there was no significant difference in Σ PFAS concentrations between Floyd and Roanoke counties, the average Σ PFAS concentration was lower in Floyd county samples than in Roanoke samples (26.98 ppt and 20.02 ppt, respectively). The highest indoor POU Σ PFAS concentration (160 ppt) was detected in a Roanoke county home; however, the overall highest Σ PFAS concentration (249.5 ppt) was sourced from an outdoor tap from a Floyd county home. This suggests that very rural areas (e.g. Floyd population ~15,000; Roanoke population ~96,000) designated by VDH as low risk still may contain significant sources of PFAS contamination. Further, there was a variation between homes in the same county in terms of PFAS detections. When considering the statistical differences between homeowner and expert collected samples, there may be several factors contributing to the differences in concentration. Because it is still possible that homeowners or experts inadvertently contaminated samples during collection or transportation, several sampling practices for homeowners will be implemented in future studies including sampling only in the mornings and all PFAS bottles for sample collection will originate from the lab completing the subsequent analysis. Not only is this practice standard in most accredited laboratories and VAHWQP but these bottles are made specifically for PFAS analysis on the LC/MS; reducing the chances of human error during bottle cleaning or residual PFAS potentially contaminating samples. Homeowners and experts also sampled the drinking water systems at different times of the day; homeowners collected samples first thing in the morning in order to collect a first draw inorganic ion sample and experts sampled mostly in the afternoons when homeowners were available for a home visit. The intra-home variability in Σ PFAS concentrations across the three POU samples could indicate that there are changes in PFAS detections that are associated with the system's water usage throughout the day.

Given the frequent detections of PFAS at the POU, it is worth noting that this study confirmed past work noting that drinking water treatment in homes supplied by private systems is still fairly uncommon. Although 65% of participants (n=18) reported using some kind of treatment device, the most common treatments were for aesthetic issues such as sediment filters or water softeners. The lack of water disinfection treatment was reflected in 30% (n=20) study homes testing positive for coliform. Although research on PFAS removal at the POU in private systems is limited, Mulhern et al. (2021) found that activated carbon filters were effective in removing emerging and legacy PFAS from homes located in areas with drinking water systems

known to be contaminated with GenX. However, only 1 out of the 20 homes sampled reported employing a carbon filter for treatment. In future efforts, it may be beneficial to recommend homeowners with high concentrations of PFAS at the POU to employ an activated carbon filter for treatment. In addition to being cited for the removal of PFAS, activated carbon filters would concurrently reduce lead and other inorganic ion concentrations in homeowner drinking water.

6.0 Future Work

Because PFAS compounds are of increasing public health and environmental concern, the amount of research regarding human- PFAS interactions will only continue to increase. The US EPA is expected to release final health advisories as well as federally enforceable MCLs for PFOA and PFOS in early 2023. The publication of interim and final health advisories as well as subsequent MCLs will allow for any future PFAS in drinking water research to be better contextualized. In addition to the updating of PFAS drinking water quality standards, the methodology and analyzation techniques for PFAS will only continue to expand. The development of analytical standards for more PFAS species will also allow researchers to better understand the actual scope of PFAS contamination as well as human-PFAS exposures. In addition to increasing the number of PFAS species that are able to be analyzed, future developments in analytics will allow for a lower detection and reporting limit for various PFAS. There will be a global demand for these standards especially if the US EPA maintains PFOS and PFOA HA limits below the current level of analytical detection. Lower detection and reporting limits will allow researchers to report more detections of PFAS in drinking water rather than only reporting samples that exceed the 70 ppt US EPA 2016 Lifetime Health Advisory.

The purpose of this project was to serve as a pilot study prior to the expansion of PFAS testing to include six counties across Virginia. These counties are also paired based on their similar geologic compositions and risk level for PFAS contamination as identified by the VDH in their 2021 municipal drinking water study. The inclusion of more POU samples through this effort will allow for a more comprehensive overview of the potential exposure to PFAS in private drinking water supplies across the state. Furthermore, the inclusion of more PFAS data will allow for increased analysis concerning mixtures and co-occurrence of short chain PFCAs and PFSAs that was documented in this study. Future studies should make a specific effort to recruit homeowners with from a greater diversity of sociodemographic and economic

backgrounds as participants in this study were wealthier and better educated than the average Virginia resident. Including more diverse households in future efforts, will allow examination of economic and sociodemographic factors as predicting factors for PFAS contamination at the POU, as well as explicit examination of the economic feasibility of existing treatment options.

Considering the frequent detection and co-detection of various short chain PFCAs and PFSAs in private and municipal drinking water supplies, it is clear there is a need for further research determining the human health risks associated with the consumption of complex mixtures of PFAAs through drinking water. Recent regulatory trends reflect new research suggesting that PFAS is harmful to human health in exceedingly small quantities (parts per quadrillion). The average drinking water sample in the present study contained a total PFAS concentration of nearly 26 ppt. Because almost all homeowners in this study indicated they drink their system's water, the potential total daily exposure of humans to PFAS in private system drinking water alone is potentially significant. Even homeowners that employed treatment devices cited as having potential for full PFAS removal such as reverse osmosis still had detectable levels of PFAS in their drinking water. The difference in PFAS mixtures inside and outside the home indicates that there are environmental and household system factors that may be contributing to PFAS concentrations at the tap. If these results are replicated, the result has a variety of implications for both municipal and private system users as the PFAS-free water leaving a water treatment plant may be influenced by a homeowner's unique plumbing.

Although the total PFAS concentrations in samples collected by expert vs homeowners was significantly different, the bias was inconsistent, and may indicate that PFAS concentrations at the tap vary throughout the day depending on water use (i.e. early morning vs afternoon), type of indoor plumbing, and presence of drinking water treatment. These differences also may reflect that PFAS concentrations vary over different days. Future work should consider attempting to quantify the incidence of PFAS at the POU over an entire day to determine if PFAS concentrations in drinking water remains relatively stable over time.

Reporting PFAS results clearly to homeowners was challenging. Given that new health advisories for PFOA and PFOS were introduced the first month of project sampling (June 2022), and that these values were below practical detection limits, it was difficult to report PFOA + PFOS detections in POU samples that were above the US EPA health advisory yet below the quantification limit for this particular effort. In addition to nuances related to detection limits, it

is worth noting that the majority of PFAS species that the US EPA recommends monitoring are not associated with any recommended limit. More than one study homeowner reached out regarding their PFAS results and simply asked how concerned their family should be about the PFAS concentrations in their water. Risk communication for emerging contaminants can be particularly difficult as human and animal health risk evaluation is often incomplete. It is also important to maintain perspective in the homeowner results letters: overestimation of potential/unknown health effects or risk related to PFAS may obscure the risks of more commonly detected and well-established health concerns such as lead, arsenic, and indicator bacteria. Future efforts should focus on examining emerging PFAS outreach efforts completed by other US states in order to continuously improve science communication in service of the public.

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Appendix A. Table of PFAS species analyzed as part of US EPA Method 533.1 and 537***

Family	Class	Group	Species	Carbon Count
Per- f l u o r o a l k y l	Perfluoroalkyl Acids (PFAAs)	Perfluoroalkyl carboxylic acids (PFCAs)	PFBA*	4
			PFPeA*	5
			PFHxA*	6
			PFHpA*	7
			PFOA**	8
			PFNA**	9
			ADONA**	10
			PFDA**	10
			PFUnDA**	11
			PFDoDA**	12
			PFTTrDA**	14
		PFTeDA**	14	
		Perfluoroalkyl sulfonic acids (PFSAs)	PFBS*	4
			PFPeS*	5
			PFHxS**	6
			FHxSA**	6
			PFHpS**	7
			PFOS**	8

			PFNS**	9
			9Cl-PF3ONS**	9
			PFDS**	10
			11Cl-PF3ONS**	11
		Fluoroalkyl ether carboxylic acid (FECAs)	HFPO-DA*	6
		Perfluoroalkyl Sulfonamides (FOSAs)	FOSA**	8
Poly- f l u o r o a l k y l	PFAA Precursors	Perfluoroalkane sulfonyl fluorides (PASFs)	MetFOSAA	11
			NetFOSAA	12
		Fluorotelomers	4:2 FTS	4
			6:2 FTS	6
			8:2 FTS	8

*A “short chain” PFAS.

**A “long chain” PFAS.

***FOSA was also analyzed as part of this method though it is not technically considered to be part of the per-or polyfluoroalkyl family.

7. In a standing glass of water, do you notice floating or settled particles?
 NO YES
 → If YES, how would you describe them? Check **all** that apply:
 White Flakes Black Specks Red-Orange Slime Brown Sediment
 Other → Please specify: _____
8. Is your water supply located **within 100 feet of the following**? Check **all** that apply:
 Septic Drain Field Home Heating Oil Storage Tank
 Pit Privy or Outhouse Pond, River, or Freshwater Stream
 Cemetery Tidal Shoreline, Estuary, or Marsh
9. Is your water supply located within a ½ mile of any of the following? Check **all** that apply:
 Landfill Golf Course Abandoned quarry, Industry, etc.
 Illegal Dump Field Crops/Nursery Farm Animal Operation
 Active Quarry Commercial Underground Storage Tank/Supply Lines (ex: gas station)
 Manufacturing/Processing Operation → Specify type: _____
10. How many people live in your household? (average number) _____
11. Have any household members who drink the water been sick to their stomach in the last 30 days?
 No. Yes, one. Yes, more than one. No one in our house drinks the water.



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Appendix C. Supplemental PFAS survey administered to participants

Virginia Household Water Quality Program

Supplementary Questionnaire
HABB1 Building- 1230 Washington St SW, Blacksburg, VA 24061
Phone: 540-231-9058 | Email: wellwater@vt.edu | Web: www.wellwater.bse.vt.edu

Sample
Number
(LAB USE
ONLY)

Principal Investigator: Dr. Leigh-Anne Krometis
Title of Study: Characterizing prevalence and risk factors of PFAS in rural private water
Sponsor: United States Geological Survey

You are invited to participate in a research study. This form includes information about the study and contact information if you have any questions.

WHAT SHOULD I KNOW?

If you decide to participate in this study, you will complete a short survey about your perceptions of your water quality and water use. The study should take approximately 10 minutes of your time. We do not anticipate any risks from completing this study. You can choose whether to be in this study or not. If you volunteer to be in this study, you may withdraw at any time without consequences of any kind. You may also refuse to answer any questions you don't want to answer and remain in the study. The investigator may withdraw you from this research if circumstances arise which warrant doing so.

CONFIDENTIALITY

Any data collected during this research study will be kept *confidential* by the researchers. Your answers will be linked back to your water quality data by your lab code, but will not be linked to your name or address. Please *do not include your name* or other identifying information in your responses that can identify you so we can be sure to keep your answers as confidential as possible.

WHO CAN I TALK TO?

If you have any questions or concerns about the research, please feel free to contact Dr. Leigh-Anne Krometis at 540-231-4372 or krometis@vt.edu. You are not waiving any legal claims, rights or remedies because of your participation in this research study. If you have questions regarding your rights as a research participant, contact the Virginia Tech HRPP office at 540-231-3732 ([HYPERLINK "mailto:irb@vt.edu" irb@vt.edu](mailto:irb@vt.edu)).

WATER CHARACTERISTICS: Please answer the following questions based on how your water is now.

1. Does your water have an unpleasant taste? yes no

If YES, how would you describe the taste? (*check all that apply*)

bitter sulfur salty metallic oily soapy other: _____

2. Does your water have an unpleasant odor? yes no

If YES, how would you describe the odor? (*check all that apply*)

rotten egg/sulfur kerosene or gas musty chemical other: _____

3. Does your water have an unnatural color or appearance? yes no

If YES, how would you describe the color or appearance? (*check all that apply*)

muddy milky black/gray tint yellow tint oily film other: _____

WATER USAGE:

4. How do you use your household water? (*check all that apply*):

- drinking
- making coffee/tea
- brushing teeth
- bathing
- other: _____
- cooking
- cleaning
- water for pets/livestock
- nothing

5. Do you purchase bottled water *for drinking*? yes no

If YES, how often (on average) do you drink bottled water?

- every day
- a few times/week
- once a week
- once a month
- once a year
- other: _____

6. Is there anywhere else you get drinking water? yes no

If YES, where else do you get drinking water? _____

7. Do you use a point of use (POU; right at the tap or under the sink) water filter for your home *drinking* water?

- yes
- no

If YES, what kind?

- faucet-mounted (i.e. a water filter connected at the kitchen tap)
- pitcher-style filter
- under the sink reverse osmosis filter
- under the sink activated carbon filter
- filter for refrigerator tap
- other: _____

SEPTIC SYSTEM

8. Do you have a septic system? yes no don't know

IF YES, proceed to question 9.

IF NO, are you connected to public sewer? yes no don't know
(then proceed to "Household Characteristics" section, i.e. Question 13)

9. Is the septic system downhill from your wellhead? yes no don't know

10. What is the approximate distance from your well to your septic system drainfield?

- <100 ft
- 101-500 ft
- 501-1000 ft

- >1001 ft
- Don't know

11. What is the approximate age of your septic system?

- <5 years
- 5-10 years
- 10-20 years
- >20 years
- Don't know

12. When was the last time you had the septic system pumped out or maintained?

- In the last year
- In the last 3 years
- In the last 5 years
- In the last 10 years
- Never
- Don't know

13. Have you ever had your septic system fail or had to have major repairs performed on it?

- yes no don't know

HOUSEHOLD CHARACTERISTICS

14. Do you rent or own your home? rent own other: _____

15. Do you know if any of the following are located within ½ mile of your home?

Airport (small or commercial)	<input type="checkbox"/> yes	<input type="checkbox"/> no	<input type="checkbox"/> unsure
Carwash	<input type="checkbox"/> yes	<input type="checkbox"/> no	<input type="checkbox"/> unsure
Waste incineration facility	<input type="checkbox"/> yes	<input type="checkbox"/> no	<input type="checkbox"/> unsure
Firefighting facility	<input type="checkbox"/> yes	<input type="checkbox"/> no	<input type="checkbox"/> unsure
Wastewater treatment plant	<input type="checkbox"/> yes	<input type="checkbox"/> no	<input type="checkbox"/> unsure
Military facility	<input type="checkbox"/> yes	<input type="checkbox"/> no	<input type="checkbox"/> unsure
Biosolids land application	<input type="checkbox"/> yes	<input type="checkbox"/> no	<input type="checkbox"/> unsure

16. How many people live in your home in each age category?

Category	Number of people
0-5 years	
6-18 years	
19-50 years	

51-65 years	
66 or older	

17. With which racial and ethnic group(s) does your household identify? (check all that apply)

- Black or African American
- Asian
- American Indian or Alaskan Native
- Native Hawaiian or Other Pacific Islander
- White
- Hispanic, Latinx or Spanish Origin
- Multiracial
- Other: _____
- I prefer not to answer

18. What is the range of your annual household income?

- Less than \$26,000
- \$27,000-52,000
- \$53,000-70,000
- \$71,000 – 97,000
- \$98,000 or above
- I prefer not to answer

19. What is the highest level of education completed by an adult in your household?

- Some high school
- High school graduate
- Some college
- College graduate
- Post college (MS, PhD)
- I prefer not to answer

HOUSEHOLD PRODUCT USE

20. Indicate how often (if ever) you or members of your home use the following products.

Cosmetics (e.g. eyeliner)	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never
Sunscreen	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never
Mosquito repellent	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never
Microwave meals	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never
Microwave popcorn	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never
Nonstick cookware	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never
Waterproofed clothing (e.g. Gore-Tex)	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never
Pest control treatment (e.g. for termites)	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never
Lawn care service	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never

4

Dryer sheets/fabric softener	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never
Aluminum foil	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never
Stainmaster treatments for carpet or upholstery	<input type="checkbox"/> daily	<input type="checkbox"/> weekly	<input type="checkbox"/> occasionally	<input type="checkbox"/> never

PRIOR PFAS AWARENESS

21. Have you heard any *national* news stories about PFAS/PFOA? yes no

22. Have you heard any *state or local* news stories about PFAS/PFOA? yes no

23. Have you seen products advertised as PFAS/PFOA-free? yes no

24. Would it influence your purchasing decision if a product is advertised as PFAS/PFOA-free? yes no

Appendix D. Sampling instructions included in homeowner drinking water sampling kits

Virginia Household Water Quality Program – SAMPLING INSTRUCTIONS

- **READ INSTRUCTIONS COMPLETELY BEFORE COLLECTING YOUR SAMPLES.**
 - **Collect your samples ONLY on the collection day you have been assigned.**
 - **Do not remove caps from sample bottles until you are ready to take each sample. Fill each of the 6 sample bottles and replace each cap one at a time as described below.**
 - **Do not write on the sample bottles.**
 - **Store samples in refrigerator or on ice following collection.**
1. In order to test for metals (lead and copper) that might be in your plumbing system, the water will need to remain in the pipes for at least 6-8 hours. **The night before your scheduled collection day, do not use any water after 10 pm.**
 2. Collect the samples first thing in the morning, **before any water has been used anywhere in the house**. Choose a cold-water faucet in the kitchen or bathroom. Be sure that the sink is clean, and that all dishes and other items are removed in order to minimize splashing. If you drink your water, and want to know about the quality of the water you drink, collect from the faucet where you obtain this water. Do not remove the aerator on the faucet.
 3. Put on the blue nitrile gloves from your sampling kit first. Because there are common substances that can interfere with our PFAS analysis **it is important to wear these gloves during the entire sample collection process**. Because you are using gloves, you will not need to wash your handles prior to collecting water for the bacteria analysis as you have done during regular VAHWQP clinics.



4. Find the bottle labeled FIRST (with an "X" on the cap) and carefully remove the cap, taking care not to touch the inside of the cap or sample bottle. Do not remove the cap from more than one bottle at a time. Do not put the cap down while collecting the sample.
5. Holding the "FIRST" bottle under the faucet, turn the water on, opening the handle as far as you normally would, and fill the bottle completely. Leave as little air as possible in the bottle, without letting the water overflow. Replace cap when full.

6. Remove the large "PFAS-1" bottle from the double bags. **Do not remove the label placed between the bags**. The special bagging strategy protects the sample from inks or other printing chemicals that may interfere with our analysis while maintaining sample identification. Fill this bottle immediately after the "FIRST" bottle and place back in the inner plastic bag.



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7. After filling these two bottles, allow the water to run at least 1 minute to flush out the pipes.
8. One at a time, fill the next three smaller sample bottles to the top, holding each cap in your hand while you fill the bottle, and taking care **not to touch the inside of the caps or the bottles**. Leave as little air as possible in each bottle. After filling each bottle, replace and tighten the cap securely. Repeat with the remaining bottles.



9. Remove the final large "PFAS ②" bottle from the double bags. Again, **do not remove the label placed between the bags**. Fill this bottle as you filled the last four, taking care not to touch the inside of the bottle or caps. After it is full, tighten the cap securely and then put it back into the inner bag. Make sure to reseal both bags.

REMEMBER! BEFORE YOU BRING YOUR SAMPLES TO THE COLLECTION POINT!

- Complete both the standard Drinking Water Clinic Questionnaire and the Supplemental PFAS Survey and bring these, with all 6 water sample bottles and these laminated sampling instructions, to the designated collection site at the designated time on your collection day. **PLEASE KEEP QUESTIONNAIRES DRY AND SEPARATE FROM BOTTLES.**

Thank you for supporting this project!

If you have any questions please contact:

1. Erin Ling, VAHWQP Director (540 - 577 - 9583)
2. Leigh-Anne Krometis, Associate Professor (540 - 553 - 1822)
3. Kathleen Hohweiler, Project Graduate Student (404 - 987 - 9990)

Appendix E. Example letter returned to participants outlining their PFAS analytical results.



COLLEGE OF ENGINEERING
COLLEGE OF AGRICULTURE AND LIFE SCIENCES
BIOLOGICAL SYSTEMS
ENGINEERING
VIRGINIA TECH.

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Blacksburg, Virginia 24061
P: (540) 231-4372 F: (540) 231-3199
krometis@vt.edu
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December 5, 2022

Dear [REDACTED]:

Thank you for participating in our United States Geological Survey (USGS) funded pilot study investigating the presence of per and poly-fluoroalkyl substances (PFAS) in private household wells (Virginia Tech study IRB #21-492). This is the *first known* effort in Virginia to systematically analyze private drinking water samples for PFAS. Your participation is essential in helping us understand best practices for collecting samples before we begin larger scale sampling efforts in your county.

*The results from your water samples are attached. All samples were directly injected onto a Ultraperformance Liquid Chromatography/Tandem Mass Spectrometry (LC/MS/MS) and analyzed for a total of 30 PFAS compounds using EPA methods 533 and 537.1. Please note that there are many different types of PFAS chemicals, and there is limited understanding of the risks that most of these pose to human health. At present, the United States Environmental Protection Agency (USEPA) has issued *final health advisories* for two specific PFAS chemicals (GenX, PFBS) and *interim health advisories* for two additional specific PFAS chemicals (PFOA, PFOS). You can read more about EPA's process for investigating PFAS impacts and potentially establishing associated regulatory limits under the Safe Drinking Water Act here: [Questions and Answers: Drinking Water Health Advisories for PFOA, PFOS, GenX Chemicals and PFBS | US EPA](#). The remaining types of PFAS we examined in your sample are recommended for monitoring, but are *not* currently associated with health advisory limits in municipal systems. Because PFAS chemical types are often proprietary, the primary sources of many of these chemicals is unknown. Your participation in our study is allowing researchers at Virginia Tech and USGS to begin to identify and understand potential relationships between typical household products, plumbing systems and PFAS in drinking water.*

Over the course of this study we collected three samples from your household tap (i.e. point of use): Kathleen Hohweiler (our graduate student) collected one sample on her initial visit, and then you collected two samples in large bottles that you brought to the Extension office. We did not observe any statistically significant difference between graduate student- and homeowner-collected samples. To be as clear as possible, we provide the maximum measured level from these three samples for comparison to the health advisory limits.

The regulatory landscape for PFAS is changing regularly for municipal water supplies and is challenging at present. It is very likely that some types of PFAS will be regulated in municipal water supplies in the near future through the Safe Drinking Water Act

(SDWA). As you know, the SDWA does *not* apply to private well water systems. Technologies to remove PFAS from drinking water are evolving and still being investigated. You can access the EPA's [Drinking Water Treatability Database \(TDB\)](#) for information on the currently best available treatment processes for controlling contaminants including PFAS.

Feel free to contact us with any questions or concerns via the email addresses below.

Sincerely,



Erin J. Ling
Sr. Extension Associate
Virginia Household Water Quality Program Coordinator
Biological Systems Engineering
wellwater@vt.edu



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Associate Professor
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Kathleen Hohweiler
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Appendix F. Example PFAS analytical results tables included with letters returned to participants

The PFAS chemicals below have USEPA *final health advisories*. If there is a trade or common name associated with the chemical, we include that in **(BOLD)** after the formal name. Health advisories are developed by USEPA to assist in monitoring contaminants that may pose a human health risk in municipal water supplies. These are not enforceable regulatory limits, though the USEPA may use these to set Safe Drinking Water Act regulations for municipal supplies in the future. These final health advisories are based on animal toxicity studies.

For each compound, we list the *maximum level of each chemical detected at your indoor tap OR list that the chemical was not detected (ND)*. Since our instruments cannot measure zero, ND basically means we didn't find any contaminant above the lowest level our instrument can detect in your sample.

For some samples, we were able to detect the contaminant, but the value was too small to be quantified (i.e. given a numeric concentration). We list these as "BQL" (*below quantification limit*). Because the quantification limit was greater than either health advisory, BQL samples can be considered below the limit.

<i>Chemical Name</i>	<i>Species</i>	<i>Final Health Advisory (ppt*)</i>	Your Sample, ppt
Perfluorobutane sulfonate (PFBS)	L-PFBS	2000	ND
Hexafluoropropylene oxide-dimer acid (GenX)	HFPO-D A	10	ND

*ppt=parts per trillion

The PFAS chemicals below have US EPA *interim health advisories*. If there is a trade or common name associated with the chemical, we include that in **(BOLD)** after the formal name. Health advisories are developed by USEPA to assist in monitoring contaminants that may pose a human health risk in municipal water supplies. These are not enforceable regulatory limits, though the USEPA may use these to set Safe Drinking Water Act regulations for municipal supplies in the future. These interim health advisories are based on human epidemiology studies.

For each compound, we list the *maximum level of each chemical detected at your indoor tap OR list that the chemical was not detected (ND)*. Since our instruments cannot measure zero, ND basically means we didn't find any contaminant above the lowest level our instrument can detect in your sample.

For some samples, we were able to detect the contaminant, but the value was too small to be quantified (i.e. given a concentration). We list these as "BQL" (*below quantification limit*). Please note that because the Interim Health Advisories for PFOS and PFOA are so low, any detection is considered above this limit.

<i>Chemical Name</i>	<i>Species</i>	<i>Interim Health Advisory (ppt*)</i>	Your Sample, ppt
Heptadecafluorooctanesulfonic acid potassium salt (PFOS)	PFOSK	0.02	ND
Perfluorooctanoic acid (PFOA)	PFOA	0.004	ND

*ppt=parts per trillion

These PFAS chemicals do not currently have US EPA recommended limits for municipal water supplies, though there are efforts to monitor levels in the environment and investigate whether limits should be established in the future. If there is a trade or common name associated with the chemical, we include that in **(BOLD)** after the formal name.

For each compound, we list the *maximum level of each chemical detected at your indoor tap OR list that the chemical was not detected (ND)*. Since our instruments cannot measure zero, ND basically means we didn't find any contaminant above the lowest level our instrument can detect in your sample.

For some samples, we were able to detect the contaminant, but the value was too small to be quantified (i.e. given a concentration). We list these as "BQL" (*below quantification limit*). For the contaminants listed below, the quantification limit was 5 ppt.

<i>Chemical Name</i>	<i>Species</i>	<i>Your Sample, ppt*</i>
Perfluorobutanoic acid	PFBA	ND
Perfluorobutane sulfonamide	FBSA	ND
4:2 Fluorotelomer sulfonic acid	4:2FTS	ND
Perfluorovaleric acid	PFPeA	ND
Perfluoropentanesulfonic acid	L-PFPeS	ND
Perfluorohexanoic acid	PFHxA	ND
Perfluorohexane sulfonamide	FHxSA	ND
Perfluorohexanesulfonic acid	PFHxSK	ND
6:2 Fluorotelomer sulfonic acid	6:2FTS	ND
9-Chlorohexadecafluoro- 3- oxanonane-1- sulfonic acid	9Cl-PF3ONS	ND
11-Chloroeicosafluoro-2-oxaundecane-1 sulfonic acid	11Cl-PF3OUdS	ND
Perfluoroheptanoic acid	PFHpA	ND
Perfluoroheptanesulfonic acid	L-PFHpS	ND
Perfluorooctane sulfonamide	FOSA	ND
8:2 Fluorotelomer sulfonic acid	8:2FTS	ND
N-methyl perfluorooctanesulfonamidoacetic acid	N-MeFOSAA	ND
N-ethyl perfluorooctanesulfonamidoacetic acid	N-EtFOSAA	ND
4,8- dioxo-3H-perfluorononanoic acid (ADONA)	NaDONA	ND
Perfluorononanoic acid	PFNA	ND
Perfluorononanesulfonic acid	L-PFNS	ND
Perfluoroundecanoic acid	PFUdA	ND
Perfluorodecanoic acid	PFDA	ND
Sodium Perfluoro-1-decanesulfonate	L-PFDS	ND
Perfluorododecanoic acid	PFDoA	ND
Perfluorotridecanoic acid	PFTTrDA	ND
Perfluorotetradecanoic acid	PFTeDA	ND

*ppt=parts per trillion

Appendix G. Inorganic ion analysis results from point of use (POU) samples collected by participants in Floyd County.

	Floyd County (n=20)			
	Metals	Maximum (ppb)	# Samples in Violation	% Homes in Violation (n=10)
MCL	As > 10 ppb	0.6	0/20	0
	Ba > 2,000 ppb	253	0/10	0
	Cd > 5 ppb	0.2	0/20	0
	Cr > 100 ppb	1.1	0/20	0
	NO ⁻³ > 10 ppm	3.25	0/10	0
	Se > 50 ppb	0.8	0/20	0
	U > 30 ppb	BD	0/10	0
TT	Cu > 1,300 ppb	1,549.30	2/20	10% 1/10
	Pb > 15 ppb	10.2	0/20	0
SMCL	Ag >100 ppb	BD	0/10	0
	Al >50 - 200 ppb	51.6	1/20	10% 1/10
	Fe > 300 ppb	1,666.52	2/20	10% 1/10
	Mn > 50 ppb	171.9	3/20	20% 2/10
	SO ₄ ⁻² > 250 ppm	10.65	0/10	0
	Zn > 5,000 ppb	4,799.10	0/20	0
	pH 6.5- 8.5	7.7	0/10	0
	Na > 20,000 ppb	47,990	2/10	20% 2/10
	Sr > 1,500 ppb	324.3	0/10	0

*BD= Below Detection

Appendix H. Inorganic ion analysis results from POU samples collected by participants in Roanoke County.

	Roanoke County (n= 20)			
	Metals	Maximum (ppb)	# Samples in Violation	% Homes in Violation (n=10)
MCL	As > 10 ppb	2.4	0/20	0
	Ba > 2,000 ppb	281.6	0/10	0
	Cd > 5 ppb	0.1	0/20	0
	Cr > 100 ppb	0.7	0/20	0
	NO ⁻³ > 10 ppm	5.77	0/10	0
	Se > 50 ppb	10.3	0/20	0
	U > 30 ppb	0.7	0/10	0
TT	Cu > 1,300 ppb	2,552.8	1/20	10% 1/10
	Pb > 15 ppb	6.2	0/20	0
SMCL	Ag >100 ppb	BD	0/10	0
	Al >50 - 200 ppb	38	0/20	0
	Fe > 300 ppb	1,536.7	2/20	20% 2/10
	Mn > 50 ppb	123.8	2/20	10% 1/10
	SO ₄ ⁻² > 250 ppm	128.15	0/10	0
	Zn > 5,000 ppb	3,750.6	0/20	0
	pH 6.5- 8.5	8	0/10	0
	Na > 20,000 ppb	196,385	7/10	70% 7/10
	Sr > 1,500 ppb	133.9	0/10	0

*BD= Below Detection

Appendix I. PFAS analysis results- POU samples in Roanoke County.

Roanoke- Inside (POU) Samples (n=30)					
	PFAS Species	Average (ppt)	Maximum Detected Value (ppt)	# Samples with Detect	% Samples with Detect
EPA Final HA	HFPO-DA (GenX) 10 ppt	ND	ND	0/30	0%
	PFBS 2,000 ppt	4.3 *	29	24/30	80%
EPA Interim HA	PFOA 0.004 ppt	8.4	20.7	7/30	23%
	PFOS 0.002 ppt	4.6 *	11.2	9/30	30%
No HA	PFBA	10.5	74.2	13/30	43%
	PFPeA	25.5	105.3	10/30	33%
	FBSA	ND	ND	0/30	0%
	PFHxA	13.6	24.9	6/30	20%
	4-2 FTS	ND	ND	0/30	0%
	L-PFPeS	3.5 *	4 *	3/30	10%
	PFHpA	5.2	9.8	7/30	23%
	NaDONA	1.6 *	2 *	6/30	20%
	FHxSA	ND	ND	0/30	0%
	PFHxSK	7.5	20.5	10/30	33%
	6-2 FTS	ND	ND	0/30	0%
	L- PFHpS	ND	ND	0/30	0%
	PFNA	ND	ND	0/30	0%
	FOSA	ND	ND	0/30	0%
	PFDA	ND	ND	0/30	0%
	8-2 FTS	ND	ND	0/30	0%
	9-Cl- PF3ONS	ND	ND	0/30	0%
	L-PFNS	ND	ND	0/30	0%
	PFUdA	ND	ND	0/30	0%
	N-MeFOSAA	ND	ND	0/30	0%
	N-EtFOSAA	ND	ND	0/30	0%
	L-PFDS	ND	ND	0/30	0%
	PFDoA	ND	ND	0/30	0%
	11-Cl-PF3OUdS	ND	ND	0/30	0%
PFTTrDA	ND	ND	0/30	0%	
PFTeDA	ND	ND	0/30	0%	

*= Below Quantification Limit (BQL), ND= Non-detect

Appendix J. PFAS analysis results- POU samples in Floyd County.

Floyd- Inside (POU) Samples (n=30)					
	PFAS Species	Average (ppt)	Maximum Detected Value (ppt)	# Samples with Detect	% Samples with Detect
EPA Final HA	HFPO-DA (GenX) 10 ppt	ND	ND	0/30	0%
	PFBS 2,000 ppt	1.07 *	2 *	13/30	43%
EPA Interim HA	PFOA 0.004 ppt	0.20 *	0.2 *	1/30	3%
	PFOS 0.002 ppt	0.55 *	0.8 *	4/30	13%
No HA	PFBA	7.6	42.9	16/30	53%
	PFPeA	8.5	26.2	4/30	13%
	FBSA	ND	ND	0/30	0%
	PFHxA	12.7	24.6	18/30	60%
	4-2 FTS	ND	ND	0/30	0%
	L-PFPeS	ND	ND	0/30	0%
	PFHpA	0.88 *	2.5 *	15/30	50%
	NaDONA	1.2 *	5.8	19/30	63%
	FHxSA	ND	ND	0/30	0%
	PFHxSK	ND	ND	0/30	0%
	6-2 FTS	17.2	23.2	9/30	30%
	L- PFHpS	ND	ND	0/30	0%
	PFNA	ND	ND	0/30	0%
	FOSA	ND	ND	0/30	0%
	PFDA	ND	ND	0/30	0%
	8-2 FTS	ND	ND	0/30	0%
	9-Cl- PF3ONS	ND	ND	0/30	0%
	L-PFNS	ND	ND	0/30	0%
	PFUdA	0.8 *	0.8 *	1/30	3%
	N-MeFOSAA	ND	ND	0/30	0%
	N-EtFOSAA	ND	ND	0/30	0%
	L-PFDS	1 *	1.8 *	2/30	7%
	PFDoA	ND	ND	0/30	0%
	11-Cl-PF3OUdS	ND	ND	0/30	0%
	PFTrDA	ND	ND	0/30	0%
	PFTeDA	7	7	1/30	3%

*= Below Quantification Limit (BQL), ND= Non-detect

Appendix K. PFAS analysis results- all POU samples collected in both counties.

Total Inside (POU) Samples (n=60)					
	PFAS Species	Average (ppt)	Maximum Detected Value (ppt)	# Samples with Detect	% Samples with Detect
EPA Final HA	HFPO-DA (GenX) 10 ppt	ND	ND	0/60	0%
	PFBS 2,000 ppt	3.2 *	29	37/60	62%
EPA Interim HA	PFOA 0.004 ppt	7.4	20.7	8/60	13%
	PFOS 0.002 ppt	3.1 *	11.2	13/60	22%
No HA	PFBA	8.9	74.2	29/60	48%
	PFPeA	20.6	105.3	14/60	23%
	FBSA	ND	ND	0/60	0%
	PFHxA	12.9	24.9	24/60	40%
	4-2 FTS	ND	ND	0/60	0%
	L-PFPeS	3.5 *	4 *	3/60	5%
	PFHpA	2.3 *	9.8	22/60	37%
	NaDONA	1.3 *	5.8	25/60	42%
	FHxSA	ND	ND	0/60	0%
	PFHxSK	7.5	20.5	10/60	17%
	6-2 FTS	17.2	23.2	9/60	15%
	L- PFHpS	ND	ND	0/60	0%
	PFNA	ND	ND	0/60	0%
	FOSA	ND	ND	0/60	0%
	PFDA	ND	ND	0/60	0%
	8-2 FTS	ND	ND	0/60	0%
	9-Cl- PF3ONS	ND	ND	0/60	0%
	L- PFNS	ND	ND	0/60	0%
	PFUdA	0.8 *	0.8 *	1/60	2%
	N-MeFOSAA	ND	ND	0/60	0%
	N-EtFOSAA	ND	ND	0/60	0%
	L-PFDS	1 *	1.8 *	2/60	3%
	PFDoA	ND	ND	0/60	0%
	11-Cl-PF3OUdS	ND	ND	0/60	0%
PFTTrDA	ND	ND	0/60	0%	
PFTeDA	7	7	1/60	2%	

*= Below Quantification Limit (BQL), ND= Non-detect

**Appendix L. PFAS analysis results from outside tap samples collected by experts in
Roanoke County**

Roanoke- Expert Outside Samples (n=10)					
	PFAS Species	Average (ppt)	Maximum Detected Value (ppt)	# Samples with Detect	% Samples with Detect
EPA Final HA	HFPO-DA (GenX) 10 ppt	ND	ND	0/10	0%
	PFBS 2,000 ppt	2.32 *	7.1	8/10	80%
EPA Interim HA	PFOA 0.004 ppt	5.25	10.1	2/10	20%
	PFOS 0.002 ppt	7.1	7.2	2/10	20%
No HA	PFBA	5.22	11.7	5/10	50%
	PFPeA	24.68	49.3	4/10	40%
	FBSA	ND	ND	0/10	0%
	PFHxA	9.83	10.9	3/10	30%
	4-2 FTS	ND	ND	0/10	0%
	L-PFPeS	2.9 *	2.9 *	1/20	10%
	PFHpA	3.57 *	6	3/10	30%
	NaDONA	2.67 *	3.1 *	3/10	30%
	FHxSA	ND	ND	0/10	0%
	PFHxSK	6.5	12.7	3/10	30%
	6-2 FTS	ND	ND	0/10	0%
	L- PFHpS	ND	ND	0/10	0%
	PFNA	ND	ND	0/10	0%
	FOSA	ND	ND	0/10	0%
	PFDA	ND	ND	0/10	0%
	8-2 FTS	ND	ND	0/10	0%
	9-Cl- PF3ONS	ND	ND	0/10	0%
	L-PFNS	ND	ND	0/10	0%
	PFUdA	3.1 *	3.1 *	1/10	10%
	N-MeFOSAA	ND	ND	0/10	0%
	N-EtFOSAA	ND	ND	0/10	0%
	L-PFDS	ND	ND	0/10	0%
	PFDoA	ND	ND	0/10	0%
	11-Cl-PF3OUdS	ND	ND	0/10	0%
	PFTTrDA	ND	ND	0/10	0%
	PFTeDA	ND	ND	0/10	0%

*= Below Quantification Limit (BQL), ND= Non-detect

Appendix M. PFAS analysis results from outside tap samples collected by experts in Floyd County

Floyd- Expert Outside Samples (n=10)					
	PFAS Species	Average (ppt)	Maximum Detected Value (ppt)	# Samples with Detect	% Samples with Detect
EPA Final HA	HFPO-DA (GenX) 10 ppt	ND	ND	0/10	0%
	PFBS 2,000 ppt	2.42 *	8.9	5/10	50%
EPA Interim HA	PFOA 0.004 ppt	0.53 *	1 *	3/10	30%
	PFOS 0.002 ppt	0.8 *	1 *	2/10	20%
No HA	PFBA	9.85	17	2/10	20%
	PFPeA	83.73	202.5	4/10	40%
	FBSA	ND	ND	0/10	0%
	PFHxA	15.83	42.6	6/10	60%
	4-2 FTS	ND	ND	0/10	0%
	L-PFPeS	ND	ND	0/10	0%
	PFHpA	1.35 *	3 *	4/10	40%
	NaDONA	3.53 *	8.2	3/10	30%
	FHxSA	ND	ND	0/10	0%
	PFHxSK	ND	ND	0/10	0%
	6-2 FTS	16.1	16.1	1/10	10%
	L- PFHpS	ND	ND	0/10	0%
	PFNA	ND	ND	0/10	0%
	FOSA	ND	ND	0/10	0%
	PFDA	ND	ND	0/10	0%
	8-2 FTS	ND	ND	0/10	0%
	9-Cl- PF3ONS	ND	ND	0/10	0%
	L-PFNS	ND	ND	0/10	0%
	PFUdA	2 *	2 *	1/10	10%
	N-MeFOSAA	ND	ND	0/10	0%
	N-EtFOSAA	ND	ND	0/10	0%
	L-PFDS	ND	ND	0/10	0%
	PFDoA	ND	ND	0/10	0%
	11-Cl-PF3OUdS	ND	ND	0/10	0%
PFTTrDA	ND	ND	0/10	0%	
PFTeDA	ND	ND	0/10	0%	

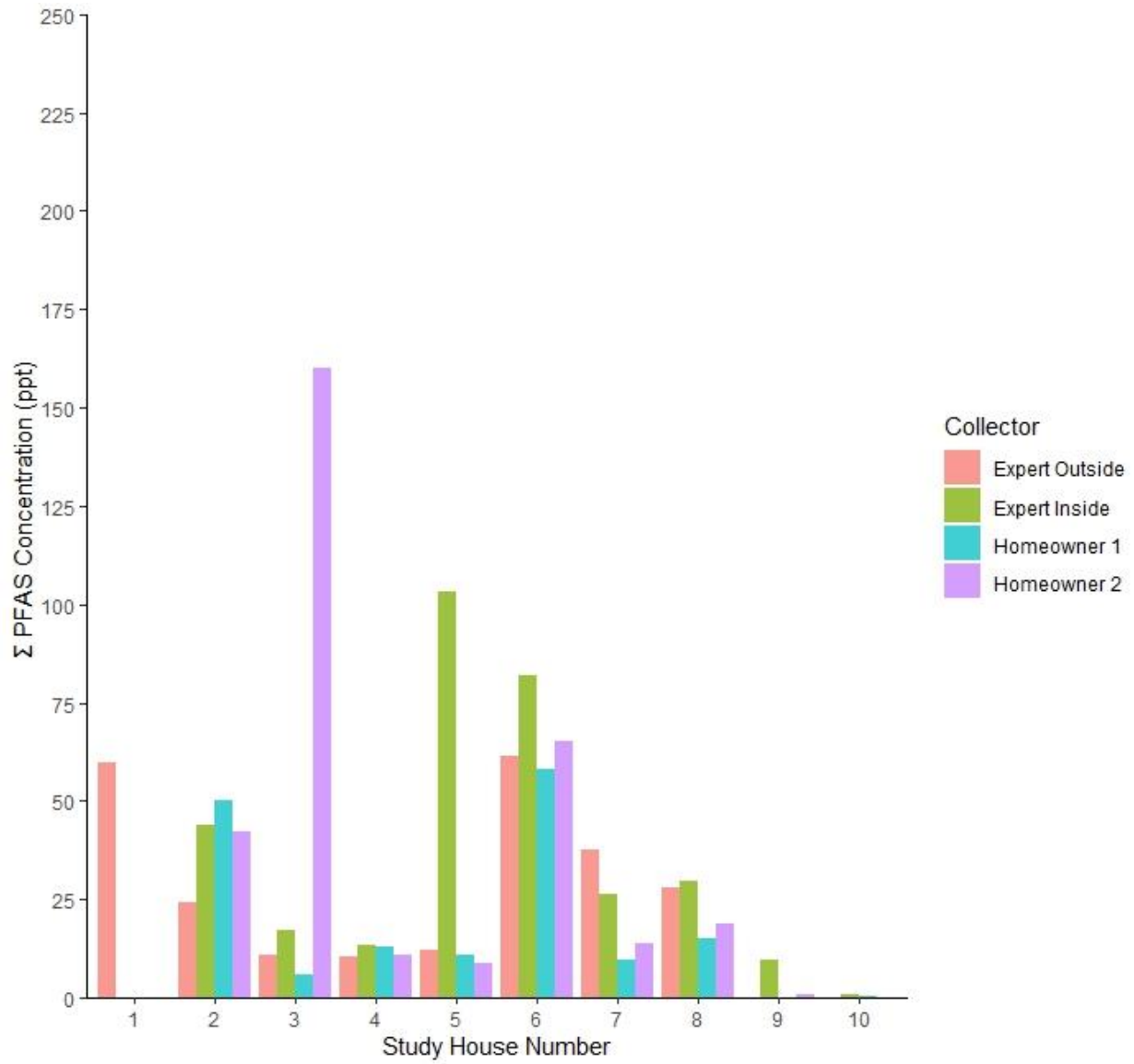
*= Below Quantification Limit (BQL), ND= Non-detect

Appendix N. PFAS analysis results from outside tap samples collected by experts in both counties

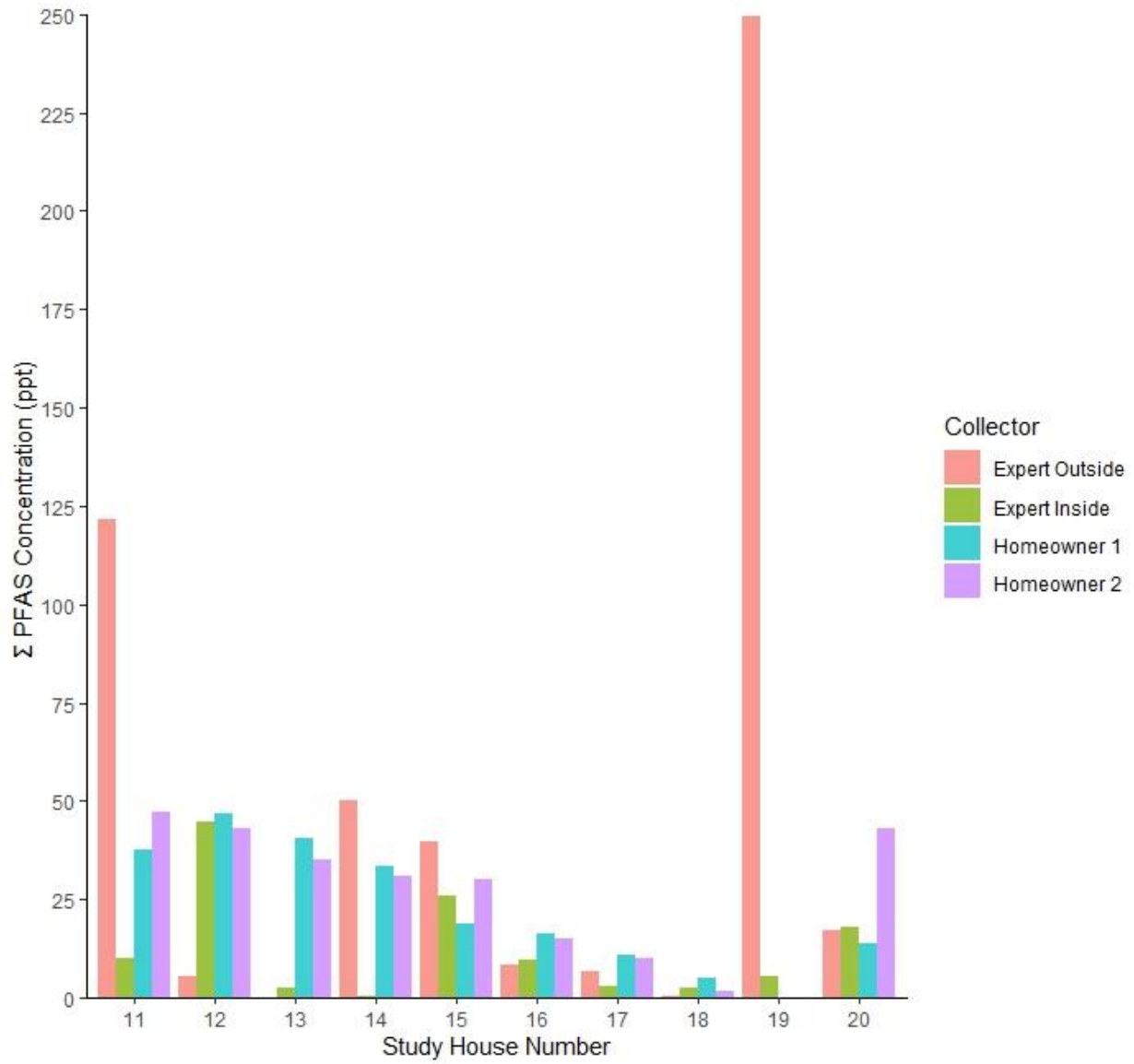
Total Expert Outside Samples (n=20)					
	PFAS Species	Average (ppt)	Maximum Detected Value (ppt)	# Samples with Detect	% Samples with Detect
EPA Final HA	HFPO-DA (GenX) 10 ppt	ND	ND	0/20	0%
	PFBS 2,000 ppt	2.36 *	8.9	13/20	65%
EPA Interim HA	PFOA 0.004 ppt	2.42 *	10.1	5/20	25%
	PFOS 0.002 ppt	3.95 *	7.2	4/20	20%
No HA	PFBA	6.54	74.2	7/20	35%
	PFPeA	54.20	202.5	8/20	40%
	FBSA	ND	ND	0/20	0%
	PFHxA	13.83	31.8	9/20	45%
	4-2 FTS	ND	ND	0/20	0%
	L-PFPeS	2.9 *	2.9 *	1/20	5%
	PFHpA	2.3 *	6	7/20	35%
	NaDONA	3.1 *	8.2	6/20	30%
	FHxSA	ND	ND	0/20	0%
	PFHxSK	6.5	10.1	3/20	15%
	6-2 FTS	16.1	16.1	1/20	5%
	L- PFHpS	ND	ND	0/20	0%
	PFNA	ND	ND	0/20	0%
	FOSA	ND	ND	0/20	0%
	PFDA	ND	ND	0/20	0%
	8-2 FTS	ND	ND	0/20	0%
	9-Cl- PF3ONS	ND	ND	0/20	0%
	L-PFNS	ND	ND	0/20	0%
	PFUdA	2.55 *	3.1 *	2/20	10%
	N-MeFOSAA	ND	ND	0/20	0%
	N-EtFOSAA	ND	ND	0/20	0%
	L-PFDS	ND	ND	0/20	0%
	PFDoA	ND	ND	0/20	0%
	11-Cl-PF3OUdS	ND	ND	0/20	0%
	PFTriDA	ND	ND	0/20	0%
	PFTeDA	ND	ND	0/20	0%

*= Below Quantification Limit (BQL), ND= Non-detect

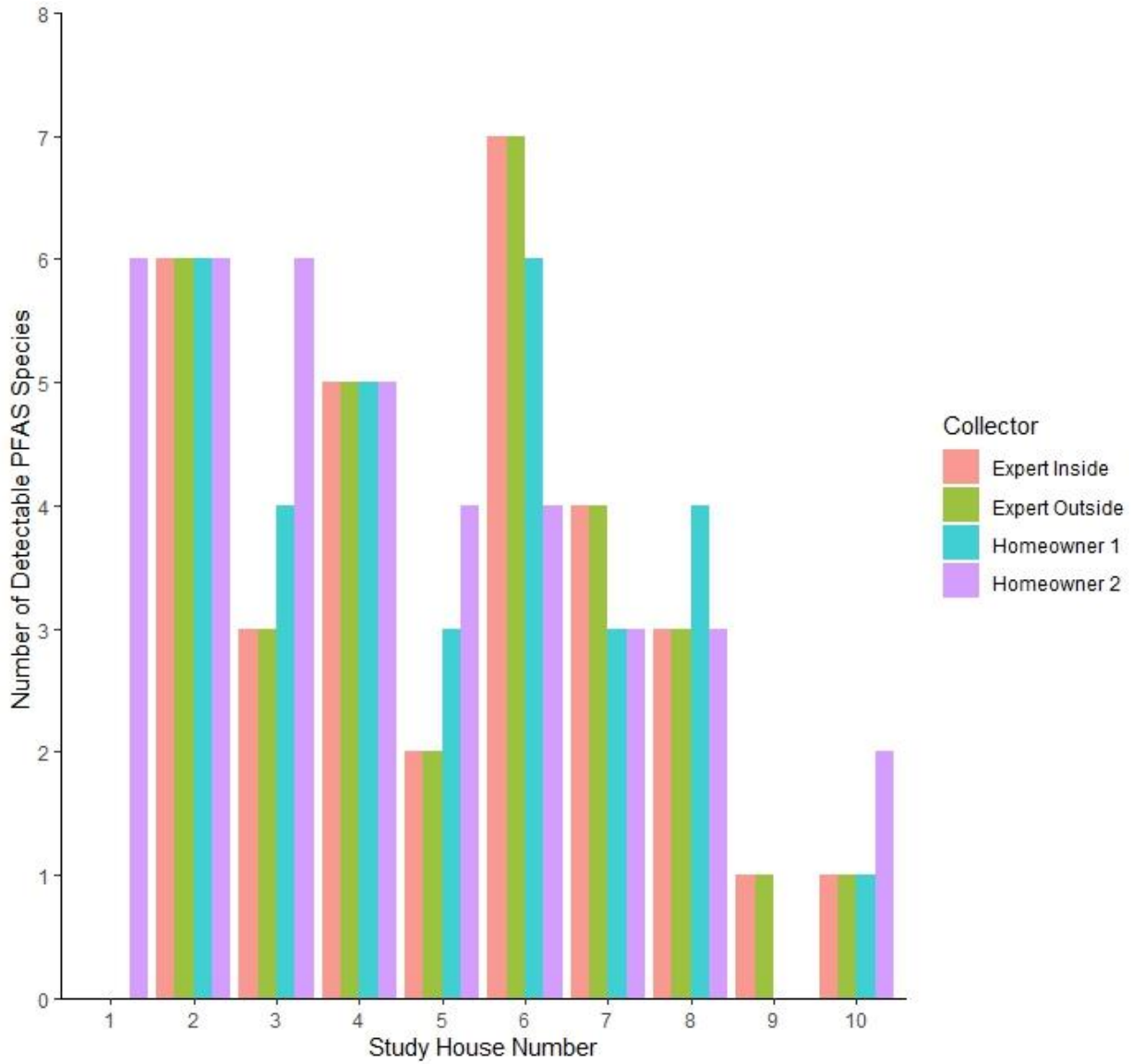
Appendix O. Σ PFAS concentrations per study household in Roanoke County



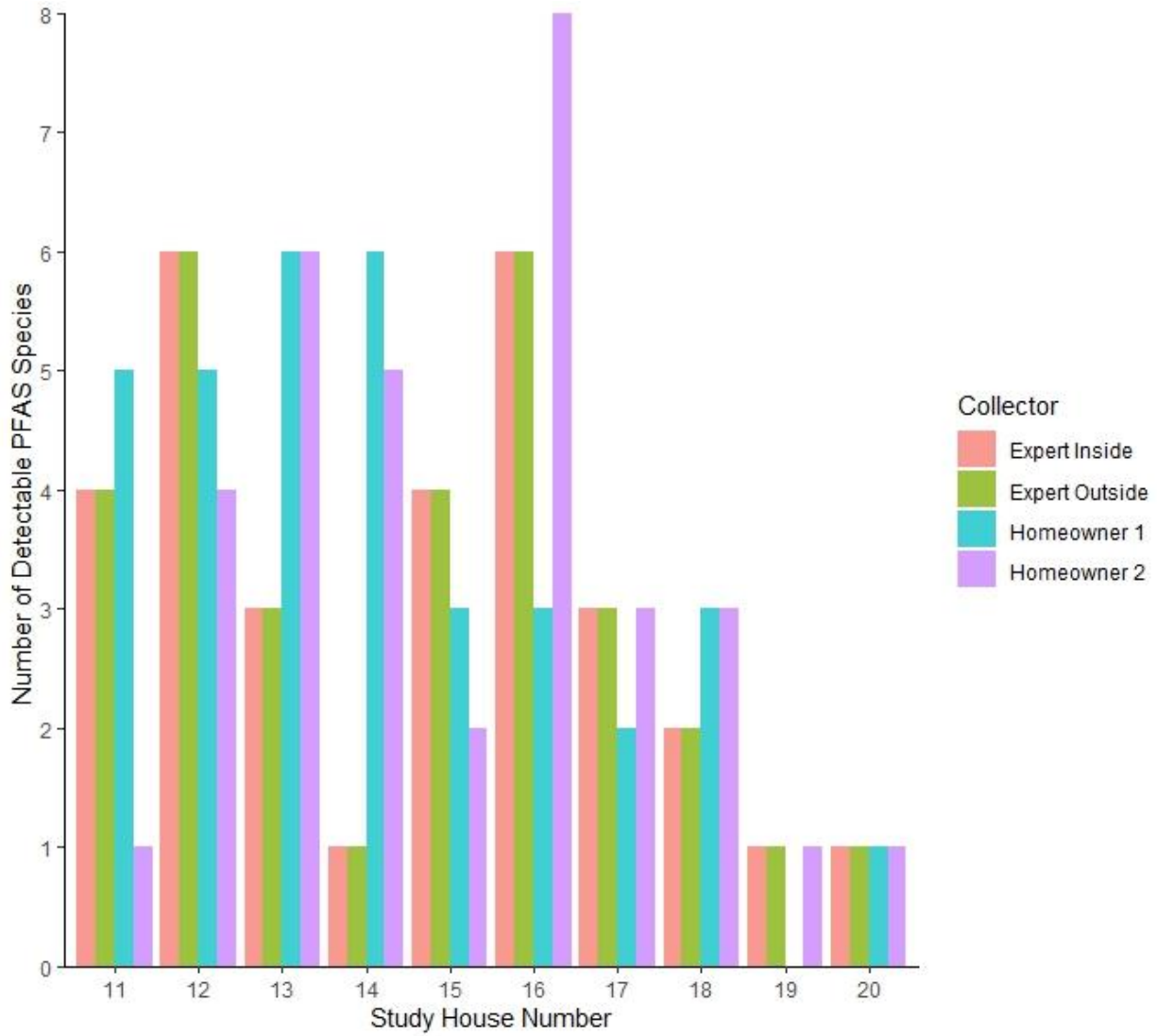
Appendix P. Σ PFAS concentrations per study household in Floyd County



Appendix Q. Number of PFAS species detected per sample per study household in Roanoke County



Appendix R. Number of PFAS species detected per sample per study household in Floyd County



Appendix S. Spearman’s correlation coefficient matrix values comparing maximum concentrations of commonly detected PFAS and inorganic ions across all POU samples

(n=60)

	Nitrate	Na	Mg	Al	Fe	Cu	Pb	PFBA	PFPeA	PFBS	PFHxA	PFHpA	ADONA	PFHxSK	PFOA	6-2FTS	PFOS
Nitrate	1	0.08	0.01	0.07	-0.37	0.39	0.1	0.1	-0.21	0.21	-0.09	0.07	-0.21	0.31	0.2	-0.15	0.02
Na	0.08	1	-0.6	-0.1	0.01	0.12	-0.2	-0.07	-0.05	0.29	-0.24	0.06	-0.39	0.49	0.11	-0.23	0.07
Mg	0.01	-0.57	1	0.17	0.09	0.11	0.1	0.24	0.3	0	0.32	-0.14	0.67	-0.55	0.01	0.32	-0.27
Al	0.07	-0.07	0.17	1	0.13	0.16	0.1	0.33	-0.1	-0.1	-0.47	-0.35	0.14	-0.15	-0.04	-0.34	-0.04
Fe	-0.37	0.01	0.09	0.13	1	-0.07	0.3	-0.08	-0.11	-0.4	0.04	-0.02	0.15	-0.32	-0.22	0.07	0.04
Cu	0.39	0.12	0.11	0.16	-0.07	1	0.6	0.22	0.12	0.35	0.06	0.27	0.15	0.25	0.01	0.03	0.17
Pb	0.08	-0.19	0.12	0.09	0.29	0.56	1	0.33	-0.21	-0.11	-0.17	-0.15	0.05	-0.28	-0.51	0.08	-0.17
PFBA	0.1	-0.07	0.24	0.33	-0.08	0.22	0.3	1	-0.2	-0.02	-0.3	-0.45	0.11	-0.34	-0.43	0.07	-0.57
PFPeA	-0.21	-0.05	0.3	-0.1	-0.11	0.12	-0.2	-0.2	1	0.55	0.56	0.23	0.6	0.17	0.23	-0.02	0.33
PFBS	0.21	0.29	0	-0.1	-0.4	0.35	-0.1	-0.02	0.55	1	0.29	0.2	0.26	0.59	0.22	0.09	0.1
PFHxA	-0.09	-0.24	0.32	-0.5	0.04	0.06	-0.2	-0.3	0.56	0.29	1	0.68	0.52	0.12	0.14	0.56	0.4
PFHpA	0.07	0.06	-0.1	-0.4	-0.02	0.27	-0.2	-0.45	0.23	0.2	0.68	1	0.1	0.49	0.4	0.42	0.73
ADONA	-0.21	-0.39	0.67	0.14	0.15	0.15	0.1	0.11	0.6	0.26	0.52	0.1	1	-0.43	-0.13	0.51	-0.12
PFHxSK	0.31	0.49	-0.6	-0.2	-0.32	0.25	-0.3	-0.34	0.17	0.59	0.12	0.49	-0.43	1	0.47	-0.28	0.61
PFOA	0.2	0.11	0.01	-0	-0.22	0.01	-0.5	-0.43	0.23	0.22	0.14	0.4	-0.13	0.47	1	-0.28	0.5
6-2 FTS	-0.15	-0.23	0.32	-0.3	0.07	0.03	0.1	0.07	-0.02	0.09	0.56	0.42	0.51	-0.28	-0.28	1	-0.21
PFOS	0.02	0.07	-0.3	-0	0.04	0.17	-0.2	-0.57	0.33	0.1	0.4	0.73	-0.12	0.61	0.5	-0.21	1