The Role of Data Analytics in Accelerating Assessments of the Impacts of Per- and Polyfluoroalkyl Substances (PFAS) in Source and Drinking Water

by

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Abstract

Per- and polyfluoroalkyl substances (PFAS) are a large, diverse, and growing family of persistent, mobile organofluorine compounds that are used for a wide variety of industrial and consumer applications, and that are ubiquitous in the environment. As the body of knowledge related to the impacts of PFAS on wildlife and human health increases, regulatory action is rapidly evolving. For example, in June 2022, the United States Environmental Protection Agency (USEPA) released new interim health advisories orders of magnitude lower than the previous advisory for two perfluoroalkyl substances: perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS). Given the complexity of PFAS issues and the costliness of PFAS data acquisition, this study investigates whether existing data are being fully exploited to address the pressing challenges related to PFAS occurrence in source and drinking water. As the understanding of environmental and human health impacts of PFAS continues to develop, and as substantial resources are allocated to acquiring PFAS occurrence data, accessible methods are needed to investigate the magnitude and extent of occurrence and exposure risks. In this study, a straightforward data analytics approach is developed using the most extensive publicly available national drinking water dataset, which features 221,831 PFAS concentration records, the USEPA's third Unregulated Contaminant Monitoring Rule round (UCMR3) data, to provide an efficient methodology for informing research directions and stakeholder decision-making. Spatiotemporal analyses were performed at the finest resolution possible to demonstrate how the approach can be used to enhance the current understanding of nationwide PFAS occurrence, visualize exposure risks, and identify geographic regions of most significant concern as new data and risk assessments

become available. The dataset was analyzed at multiple spatial scales (national, regional, state, individual water systems, and sample collection points). To the best of the author's knowledge, the results reported herein are the first to 1) spatially analyze the results of the only currently existing nationwide database of PFAS in drinking water at the individual public water system (PWS) level, 2) evaluate nationwide PFAS exposure through drinking water using the USEPA's 2022 interim lifetime health advisories for PFOA and PFOS, 3) evaluate the impact of database structure and varying data aggregation methods on analysis outcomes, and 4) demonstrate the utility of existing and pending UCMR data to investigate trends in PFAS occurrence in source water. The results will allow scientists, stakeholders, and regulatory agencies to identify geographic hotspots with the most significant levels of PFAS contamination; better understand nuances in database structure that affect the current understanding of exposure risks; and initiate efficient data management and analysis methods to more fully leverage large quantities of both currently available and future PFAS data.

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List of Abbreviations

ADEM	Alabama Department of Environmental Management
AFFF	Aqueous film forming foams
AWWA	American Water Works Association
CCR	Consumer confidence report
CDC	U.S. Centers for Disease Control and Prevention
EP	Entry point to drinking water distribution system
GW	Groundwater
HUC	Hydrologic unit code
LC-MS/MS	Liquid chromatography tandem mass spectrometry
LHA	Lifetime Health Advisory
MCL	Maximum contaminant level
NCOD	National Contaminant Occurrence Database
OECD	United Nations Organization of Economic Corporation and Development
PFAS	Per- and polyfluoroalkyl substances
PFBS	Perfluorobutanesulfonic acid
PFCA	Perfluoroalkylcarboxylic acid
PFHpA	Perfluoroheptanoic acid
PFHxS	Perfluorohexanesulfonic acid
PFNA	Perfluorononanoic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctanesulfonic acid
PFSA	Perfluoroalkylsulfonic acid
PWS	Public water system
RO	Reverse osmosis
SDWA	Safe Drinking Water Act
SW	Surface water
UCMR3	Third Unregulated Contaminant Monitoring Rule (2013-2015)
UCMR5	Fifth Unregulated Contaminant Monitoring Rule (2023-2025)
USEPA	United States Environmental Protection Agency
WTP	Water treatment plant
WWTP	Wastewater treatment plant

Chapter 1. Introduction

1.1 Background

Per- and polyfluoroalkyl substances (PFAS) are a large and growing family of anthropogenic organofluorine compounds with unique properties that are leveraged for a multitude of consumer and industrial applications, but that have become a pressing and increasingly complex issue within the water community and for the public at large (Blum et al., 2015; Kwiatkowski et al., 2020; Speth et al., 2022; Wang et al., 2017). Some PFAS are well-studied with known negative health effects including cancer, immune dysfunction, and endocrine disruption (Blum et al., 2015; Wang et al., 2017; Xiao, 2017). The effects of many other PFAS, however, remain largely unknown (Bell et al., 2021; Fenton et al., 2021). Drinking water has been identified as a substantial source of PFAS exposure for many populations, particularly in communities with known PFAS contamination (Domingo & Nadal, 2019; Sunderland et al., 2018). Until the present, there have been no enforceable drinking water standards for any PFAS in the United States; however, federal regulation of two PFAS is imminent: perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS). In March 2021, the United States Environmental Protection Agency (USEPA) published a final determination to regulate PFOA and PFOS (U.S. EPA, 2021a). The proposed regulation is anticipated by the end of 2022, with the final regulation expected in the fall of 2023 after consideration of public comments (U.S. EPA, 2021d).

Regulatory action is increasing globally as negative health outcomes related to PFAS exposure are better understood; however, replacing well-studied PFAS with negative impacts on humans and wildlife, commonly referred to as “legacy” PFAS, with structurally similar PFAS for which less information regarding effects on humans and the environment exists, is a common practice of concern (Blum et al., 2015; Kotthoff & Bücking, 2018; Scheringer et al., 2014; Sunderland et al., 2018; Wang et al., 2017). In 2017, the total number of PFAS in existence was estimated to be 3,000 (Wang et al., 2017). Recent estimates of the total number of PFAS in existence exceed 12,000 (Salvatore et al., 2022; U.S. EPA, n.d.-b). As the total number of PFAS on the global market has substantially increased, health advisory levels have substantially decreased. In 2009, the USEPA issued a provisional health advisory for PFOA and PFOS of 400 and 200 ng L^{-1}, respectively. In 2016, the USEPA updated this guidance with a non-enforceable interim lifetime drinking water health advisory (LHA) for PFOA and PFOS combined: 70 ng L^{-1} for the sum of the two concentrations in a given water supply (U.S. EPA, 2016b). In June 2022, new interim LHAs were established for PFOA and PFOS individually: 0.004 ng L^{-1} and 0.02 ng L^{-1}, respectively (U.S. EPA, 2022f).

For many reasons, PFAS present unique environmental, human health, societal, economic, engineering, and research challenges. PFAS pose substantial challenges to drinking water utilities because they necessitate advanced, costly treatment technologies and prevent the use of traditional approaches for handling waste streams (Burns et al., 2020). For example, due to significant PFAS contamination in the Cape Fear River watershed, Brunswick County, North Carolina is spending more than $167 million on a reverse osmosis (RO) treatment plant, and the affected public utility spent $46 million on granular activated carbon filters (Cordner et al., 2021). In a survey of approximately 300 stakeholders in the North American water and wastewater industry, 49% cited PFAS a chief contaminant issue of concern, and 57% cited regulatory uncertainty as the limiting factor for addressing PFAS in water supplies (Burns et al., 2020). In a testimony on behalf of the American Water Works Association (AWWA) to the U.S. Senate's Committee on Environment and Public Works on the topic of PFAS, it was noted that "drinking water utilities and state environmental agencies need to know where to focus monitoring efforts to understand what risks may be in source waters" (Mehan, 2021). While data are currently limited regarding where PFAS compounds have been produced and in what volumes (Mehan, 2021; Salvatore et al., 2022), gleaning as much information as possible from existing, publicly available data can maximize the current understanding of PFAS occurrence and exposure risks in source and drinking water.

In 2012, the USEPA included six PFAS in their third Unregulated Contaminant Monitoring Rule (UCMR3). The UCMR is used by the USEPA to collect data for contaminants suspected to be present in drinking water with no existing health-based standards set under the Safe Drinking Water Act (SDWA) (U.S. EPA, 2017). Resulting occurrence data are intended to be used to evaluate populations being exposed to contaminants of emerging concern and at what levels of exposure; and ultimately support decision-making regarding the regulation of contaminants in order to protect public health (U.S. EPA, 2021b). The data are stored in a publicly available database, the National Contaminant Occurrence Database (NCOD) to satisfy the requirements of the SDWA (U.S. EPA, 2021b, 2021c). The UCMR3 mandated all public water systems (PWSs) in the U.S. serving drinking water to at least 10,000 consumers to collect treated drinking water samples from each entry point to their distribution network (EP) for delivery to customers (U.S. EPA, 2012b). In December 2021, the USEPA issued the fifth Unregulated Contaminant Monitoring Rule (UCMR5) which, in comparison to UCMR3, reduced the minimum number of consumers requiring a PWS to monitor for PFAS (at least 3,300), increased the number of PFAS to be monitored from six to twenty-nine, and decreased the minimum concentrations requiring reporting in the publicly available database (U.S. EPA, 2021e). These monitoring efforts are extremely costly. The initial cost projection of the implementation of the UCMR3 was $17.45 million annually from 2012-2016, i.e., $104.7 million in total (U.S. EPA, 2012b). The UCMR5 is projected to cost $20.8 million annually from 2022-2026 ($124.8 million in total).

Scientists and stakeholders are keenly interested in understanding the global distribution of PFAS and the risks associated with exposure (Crimi et al., 2022; Harclerode et al., 2021; Mehan, 2021), but many challenges hamper scientific progress in PFAS research (Guelfo et al., 2018; Speth et al., 2022; Wang et al., 2017). Although concentrated efforts to improve the current understanding of PFAS occurrence and health effects are underway across the globe (Blum et al., 2015; Naidu et al., 2020; U.S. EPA, 2021d), the list of PFAS relevant to human health and the environment is continually increasing (Bell et al., 2021; Gebbink et al., 2016; Kwiatkowski et al., 2020; Liu et al., 2019; Munoz et al., 2019; Wang et al., 2017); and there are currently no reference standards available to quantify many of them with liquid chromatography tandem mass spectrometry (LC-MS/MS) analyses (Tian & Sun, 2019). Broad implementation of targeted PFAS analyses is further hampered by the cost and time required for analysis and an incomplete understanding of which of the thousands of PFAS should be prioritized in monitoring efforts (Guelfo et al., 2018; Kotthoff & Bücking, 2018). The rapidly evolving body of knowledge related PFAS presents its own challenge. As noted in a 2022 roundtable interview amongst PFAS experts, “It is a major challenge simply to keep up with the everchanging information as it’s generated” (Crimi et al., 2022). Approaches are needed to rapidly process new information related to PFAS occurrence and exposure risks. This study focuses on the strategic utilization of existing PFAS data as an avenue to accelerate assessments of the impacts of PFAS occurrence in source and drinking water, better understand exposure risks, guide investigative and remediation efforts, design relevant studies and experiments, and inform regulatory decisions.

1.2 Research objectives

Given the increasingly complex nature of PFAS problems, the resources required to address pressing PFAS challenges, and the expensive and intensive data acquisition efforts underway, the following research questions were posed. The overall objectives of this research are to answer these key questions.

1) Have the existing nationwide drinking water monitoring data been fully exploited to aid in addressing the increasingly pressing need to understand PFAS occurrence and exposure risks? If not, how can the utilization of these data be expanded?
2) Are there accessible methodologies in place to efficiently process and analyze pending, larger amounts of nationwide data and produce meaningful information as the understanding of the risks of PFAS exposure continues to evolve?
3) Do previous and future data management structures provide adequate information to accurately identify populations exposed to PFAS, which PFAS, and at what concentrations they are exposed?

Investigation of the above research questions resulted in the identification and execution of the following objectives: (1) review uses of the most extensive PFAS drinking water monitoring effort to date (the USEPA UCMR3) in the scientific literature and the methods applied to analyze PFAS occurrence and risks; (2) develop an efficient, accessible data analysis methodology that allows for existing and future PFAS monitoring data to be analyzed at the finest spatial resolution possible (the PWS-level); (3) analyze a subset of the UCMR3 data at the distribution system entry point (EP) level to refine the current understanding of PFAS occurrence and exposure risks; (4) investigate methods to expand utilization of existing and pending drinking water monitoring data; and (5) identify weaknesses in current data management structures that limit the ability of scientists and engineers to fully exploit the information acquired by expensive and intensive data acquisition efforts.

1.3 Expected outcomes

The intended outcomes of this research include the development and application of a straightforward, accessible data analysis methodology that would foster accelerated progress in understanding and addressing the problems associated with PFAS in source and drinking water amongst the scientific community, stakeholders, and the public. In the scientific community, efficient methods for processing and extracting meaning from existing and future PFAS data are needed to inform research directions. This work aims to leverage existing data produced by the only nationwide routine drinking water PFAS monitoring effort to date to its fullest extent, which will provide new information from existing data, as well as an approach for expediting analyses of additional state-wide datasets and pending nationwide monitoring data. Additionally, by investigating the effects of varying data management, aggregation, and analysis approaches, this will aid scientists in developing a clearer understanding of conclusions reached in previous PFAS investigations and their implications.

Stakeholders include regulatory agencies at federal and state levels; municipalities and utilities; fluorochemical manufacturers; and the civil and environmental engineering community. By providing resources and recommendations for maximizing the use of costly PFAS monitoring data, this research will greater assist federal and state agencies in strategic decision-making in the development of feasible action plans to address PFAS contamination in drinking water and environmental media. Spatially analyzing existing nationwide PFAS data will allow under-resourced municipalities and utilities anticipate the impacts of the implementation of new maximum contaminant levels (MCLs) of PFAS at the federal level. This research will also provide fluorochemical manufacturers with a resource for better understanding PFAS release impacts and associated liabilities. The framework and spatial analyses will be a resource for civil and environmental engineers by providing those serving in the public sector a better understanding of the risks of PFAS contamination in the source water of their utilities, and those in the design and construction industries an opportunity to identify market expansion opportunities in PFAS remediation and treatment. The public will benefit from the results of this work as well, as awareness and confusion regarding effects of PFAS on wildlife and human health increase. Overall, applying systematic data analytics approaches to advance the state of the science related to PFAS occurrence in source and drinking water will allow scientists, stakeholders, and the public to become better informed regarding the magnitude and extent of PFAS problems.

1.4 Organization of dissertation

This dissertation is divided into five chapters that describe the steps taken to meet the defined objectives and their results. Chapter 2 provides further information regarding the evolution of PFAS occurrence as a global environmental and human health issue; details the challenges related to assessing and managing PFAS; explains how exploiting existing PFAS monitoring data to its full extent presents a unique opportunity to address these challenges; and reviews utilization of the USEPA UCMR3 data (Objective 1). The focus of Chapter 3 is the development of a methodology for querying, aggregating, and analyzing both currently available and future UCMR PFAS drinking water data to maximize understanding of PFAS occurrence and risks (Objective 2). The methodology is applied to spatially analyze the UCMR3 occurrence data for each PFAS monitored at the finest resolution possible (the PWS-level). Chapter 4 demonstrates the utility of the approach developed in Chapter 3 for understanding PFAS exposure risks in a changing regulatory climate by analyzing the UCMR3 dataset in terms of past and present health benchmarks. In this chapter, the data are also analyzed at the distribution system EP-level to highlight nuances in data structure, which affect various analysis outcomes and the current understanding of exposure risks (Objective 3). In this chapter, the need for clear communication of data management and analysis practices in PFAS research is also demonstrated. Additionally, this chapter explores the application of the UCMR data for guiding investigations of PFAS in source water (Objective 4). Chapter 5 summarizes the impact of this work; details aspects of current data management structures that limit full exploitation of expensive and intensive data acquisition efforts (Objective 5); and recommends future research directions.

Chapter 2. Literature Review

2.1 Introduction

While the term “fluorinated substances” generically describes a vast number of organic and inorganic substances containing at least one fluorine atom that exhibit a wide variety of physical, chemical, and biological properties (Banks et al., 2013), PFAS are a distinct, albeit large, subset of highly fluorinated aliphatic substances, many of which share common properties. Notionally derived from hydrocarbons, the distinguishing characteristic of PFAS is at least one carbon atom on which all of hydrogen atoms have been replaced by fluorine atoms, yielding the perfluoroalkyl moiety $C_nF_{n2+1}^-$ (Buck et al., 2011; OECD/UNEP Global PFC Group, 2013). The bond between carbon and fluorine is among the strongest bonds in organic chemistry (O’hagan, 2008). Due to the characteristic perfluoroalkyl moiety, PFAS can exhibit unique characteristics, such as chemical and thermal stability; hydrophobicity (water repellence); lipophobicity (oil repellence); and the ability to lower aqueous surface tension (Buck et al., 2011; Smart, 1994). These properties make PFAS useful for a wide variety of polymer and surfactant applications (Buck et al., 2011; Wang et al., 2017).

Uses of PFAS range in scale from personal care products and food packaging to industrial applications in aerospace, construction, and electronics (Brennan et al., 2021; Teng, 2012). The properties that make these chemicals useful for so many aspects of modern life, such as their resistance to degradation, present significant environmental challenges. PFAS have become ubiquitous in the environment, having been found in various environmental media worldwide, from highly industrial areas to remote regions like Antarctica (Brendel et al., 2018; Llorca et al., 2012). In addition to their ubiquity in environmental media, these compounds have also been detected in a variety of living organisms including humans (Ahrens & Bundschuh, 2014; Ankley et al., 2021). The growing understanding of the environmental persistence and biological impacts of PFAS has led to global concern regarding the long-term consequences of PFAS exposure on human health and wildlife (Brennan et al., 2021), as well as substantial shifts in the types of PFAS being produced (Blum et al., 2015; De Silva et al., 2021; Scheringer et al., 2014). This chapter provides an overview of the evolution of PFAS as a global environmental and human health issue and further details the factors contributing to the complexity of assessing and managing PFAS. Priority needs for the water industry to address PFAS effectively and economically are identified, and the utility of data analytics for accelerating progress in meeting those needs is explored.

2.2 Evolution of per- and polyfluoroalkyl substances (PFAS) as a global issue

2.2.1. Origins and applications

The wide-reaching fluoropolymer industry began in 1938 with the discovery of polytetrafluoroethylene (PTFE) at DuPont (Plunkett, 1986; Teng, 2012). PTFE became commercially available with DuPont's trademark Teflon in 1947 (Teng, 2012), and the 3M Company began producing perfluorochemicals in 1949 (Paul et al., 2009). Since that time, PFAS have proliferated in both number and applications (De Silva et al., 2021; Teng, 2012; Wang et al., 2017). Publications on fluorine-containing products numbered 536,000 from 1989-1998; and of these, 144,720 were patents (Johns & Stead, 2000). Production and use grew substantially until the 1990s, with uses including inks, varnishes, waxes, fire-fighting foams, metal plating and cleaning, lubricants, and water and oil repellents for leather, paper, and textiles (Paul et al., 2009). A review of the development of the fluoropolymer industry indicated that the market growth for PTFE alone was 3%-5% per year for three decades beginning in the 1980s, with annual worldwide consumption of PTFE over 55,000 tons by the late 1990s and consumption nearly doubling by 2012 (Teng, 2012).

The impact of PFAS on modern life is wide-reaching, with uses in healthcare and emergency response; construction; chemical and pharmaceutical manufacturing; aerospace and defense; outdoor apparel and equipment; and cosmetics and personal care products (American Chemistry Council, 2020; Glüge et al., 2020). Because PFAS are prevalent in many aspects of modern life, their economic value is significant (Ahrens & Bundschuh, 2014). For example, the stain repellents market, one of many markets that relies heavily on PFAS, was valued at nearly \$1 billion annually in 2006 (Renner, 2006). In 2018, an association of the world's leading fluoropolymer manufacturers estimated \$2 billion in direct fluoropolymer sales in the United States alone (Wood Environment & Infrastructure Solutions UK Limited, 2020). The proliferation of PFAS presents a major societal challenge because, on the one hand, this family of chemicals is among the most innovative developments in materials chemistry with many applications in modern society, some of which are essential (Bowman, 2015; De Silva et al., 2021; Johns & Stead, 2000). On the other hand, there are substantial concerns regarding the ecological and human health impacts of PFAS exposures due to decades of widespread global use, high persistence, and mobility (De Silva et al., 2021; Kwiatkowski et al., 2020).

2.2.2. Evolving understanding of health effects

Most research on PFAS occurrence and exposure risks began in the early 2000s; however, the 3M Company began studying health effects of PFAS exposures in animals and humans before 1980 (Sunderland et al., 2018). One acute animal toxicity study associated with exposure to legacy PFAS, a 90-day rhesus monkey study, had to be aborted after monkeys in all treatment groups died after 20 days (Goldenthal et al., 1978a). Reductions in total cholesterol, increased liver weight, and immune system toxicity were observed in later monkey studies using lower doses (Goldenthal et al., 1978b). Health surveillance of 3M workers yielded inconsistent results; however, one study found that exposure to a legacy PFAS, perfluorooctanoic acid (PFOA), may significantly alter male reproductive hormones and leukocyte counts (Gilliland, 1992).

In 2002, in its hazard assessment for the legacy PFAS perfluorooctanesulfonic acid (PFOS), the United Nations Organization of Economic Corporation and Development (OECD) determined PFOS to cause liver and thyroid cancer in rats (OECD, 2002). Moreover, the assessment indicated that PFOS exposure was epidemiologically linked to bladder cancer in humans, and ultimately concluded that PFOS is “persistent, bioaccumulative, and toxic to mammalian species” (OECD, 2002). In an investigation performed by the U.S. Centers for Disease Control and Prevention (CDC) using the blood serum of a representative sample of the general American population (collected from 2003-2004, 2,094 participants), PFOS was detected in 99.9% of American blood serum samples (Calafat et al., 2007), a discovery that further drove human health concerns around PFAS production and use (Brennan et al., 2021). A decade later, a review of epidemiological literature suggested links between human PFOS exposure and higher cholesterol, impaired glucose metabolism, increased body mass index, impaired thyroid function, infertility, and attention deficit hyperactivity disorder (Saikat et al., 2013). In 2014, a document summarizing scientific concerns and potential for harm of PFAS (Blum et al., 2015), was presented at the Dioxin Symposium in Madrid, Spain and signed by more than 250 scientists from 38 countries (Green Science Policy Institute, n.d.). The statement reported significant or suggestive associations between specific legacy PFAS and adverse outcomes in the growing body of epidemiological evidence (Blum et al., 2015). These outcomes include testicular and kidney cancers, liver malfunction, hypothyroidism, high cholesterol, ulcerative colitis, lower birth weight and size, obesity, decreased immune response to vaccines, reduced hormone levels, and delayed puberty (Blum et al., 2015).

Experimental results from rodent studies can be challenging to translate directly to human health impacts because of differences in peroxisome proliferation expression, one of the main mechanisms of PFAS toxicity (Brase et al., 2021; Palmer et al., 1998; Sunderland et al., 2018). To date, the most comprehensive longitudinal evidence for adverse human health effects associated with PFAS exposure is from the population living near the West Virginia DuPont Washington Works fluorotelomer plant through the C8 Health Project (Sunderland et al., 2018). The project found probable links between PFOA and high cholesterol, thyroid disease, pregnancy-induced hypertension, ulcerative colitis, and kidney and testicular cancer. Such health concerns have led to voluntary and regulatory phase-outs of PFOA and PFOS; however, these substances are being replaced by other fluorinated alternatives with unknown health effects (Blum et al., 2015; Scheringer et al., 2014; Wang et al., 2017). No reference standards are currently available to quantify many of these novel PFAS using LC-MS/MS analysis (Tian & Sun, 2019). Quantitation of PFAS with available standards is costly and time-consuming; and an incomplete understanding of which PFAS should be prioritized further complicate efforts to understand occurrence and impacts (Guelfo et al., 2018; Kotthoff & Bücking, 2018). For these and other reasons, assessing and managing these additional PFAS require substantial research and regulatory efforts; and it is technically and financially challenging to identify and reverse environmental and human exposure to this expanding family of chemicals (Wang et al., 2017).

Thus far, most human exposure assessments have focused on two to five individual PFAS (De Silva et al., 2021). Human health risk assessments of PFAS exposure are complicated by a number of factors, which include a lack of clear understanding of which PFAS may be relevant for potential human health impacts, relatively sparse information on PFAS toxicity and human exposure, and the fact that most humans are exposed to an unknown mixture of PFAS (Anderson et al., 2022). Further, toxicity test results often lack agreement among assays in animals and observations in humans, and extrapolation from animal data to human relevance is highly uncertain (Anderson et al., 2022).

2.2.3. Health guidance and regulatory action

International bodies, individual countries, states, and local areas have undertaken efforts to limit PFAS production and human exposure by setting restrictions on the manufacturing and trade of PFAS, setting maximum permissible levels of PFAS in food and drinking water, and regulating the disposal of products that contain PFAS (Brennan et al., 2021). However, there is not a consistent regulatory structure for PFAS, there is disagreement on appropriate level of regulation, and standards vary widely (Brennan et al., 2021). Due to common physicochemical, environmental, and toxicological properties of many PFAS, some scientists have recommended a class-based approach for regulating and managing PFAS (Cousins et al., 2020; Kwiatkowski et al., 2020). Others, however, argue that the physicochemical behavior of PFAS as one class has been overgeneralized based on assessments of a relatively small number of substances, noting that further research is needed prior to implementing class-based regulation of thousands of chemical compounds (Singh & Papanastasiou, 2021). In fact, there is some disagreement among scientists, stakeholders, and regulatory agencies about what constitutes the classification of a chemical as a PFAS to begin with, as broader definitions include a number of fluorochemicals that would otherwise not be classified as PFAS, and for which there is currently less evidence of persistence, bioaccumulation, and toxicity (Singh & Papanastasiou, 2021). The USEPA has noted "there is no precisely clear definition of what constitutes a PFAS substance given the inclusion of partially fluorinated substances, polymers, and ill-defined reaction products" (U.S. EPA, n.d.-b). In discussing the factors complicating the current understanding of PFAS human health risk assessments, Anderson et al. included the lack of consensus regarding the definition of what is and is not a substance within the PFAS family (Anderson et al., 2022).

There is general agreement that ingestion is the largest source of PFAS exposure (as opposed to inhalation or dermal contact), but the relative importance of these exposure pathways varies across demographic groups and populations (Domingo & Nadal, 2019; Sunderland et al., 2018). In communities near sites with known PFAS contamination, drinking water has been reported to account for up to 75% of total PFAS exposure. PFAS enter the environment (and consequently, drinking water systems) through both direct and diffuse emissions. Direct emissions include those of fluorochemical manufacturing facilities; manufacturing facilities where PFAS are used; aqueous film forming foams (AFFF) used at airports and military bases for firefighting and training activities; wastewater treatment plants (WWTPs); and landfills (Banzhaf et al., 2017; De Silva et al., 2021; Hu et al., 2016; Sunderland et al., 2018). Although fluorochemical manufacturing sites are relatively few in number, these sites are responsible for a large proportion of the global emissions of some classes of PFAS, and high-volume emissions from these facilities can impact large geographic areas (De Silva et al., 2021). Efforts and regulatory actions to eliminate non-essential use of PFAS, and to reduce discharges of PFAS at point sources, are underway worldwide (Brennan et al., 2021; U.S. EPA, 2021d). However, PFAS that are ubiquitously found in the environment and drinking water must be addressed (Crimi et al., 2022).

2.3 Data analytics as a resource for effective PFAS response

Compared to other PFAS research methods, few peer-reviewed studies have utilized "big data" (Klievink et al., 2016) and geospatial methodologies (Tim, 1995) to answer PFAS-specific research questions at varied geographic scales, despite thousands of PFAS-related peer-reviewed articles being published since 2002 (Wang et al., 2017). As PFAS detection and quantitation methods improve, larger quantities of data are becoming more widely available to scientists and stakeholders. As noted previously, the USEPA UCMR3 mandated all PWSs in the U.S. serving drinking water to at least 10,000 consumers collect treated drinking water samples from each entry point to their distribution network (U.S. EPA, 2012b). In comparison, the UCMR5 reduced the minimum number of consumers requiring a PWS to monitor for PFAS to 3,300 (subject to available USEPA appropriations); increased the number of PFAS to be monitored from six to twenty-nine; and substantially decreased the minimum concentrations requiring reporting in the publicly available database (U.S. EPA, 2021e).

As pressure to address PFAS contamination increases across the water sector, the increasing availability of large PFAS datasets provides a unique opportunity to better understand the occurrence, behavior, fate, and effects of these chemicals in freshwater and drinking water systems. The UCMR3, which began in 2013 and ended in 2016, is the most extensive PFAS drinking water monitoring efforts to date, both in geographic reach and in quantity of data produced. The data provide information regarding mixtures of PFAS from PWSs of various sizes using various source water types (surface water, groundwater, groundwater under the influence of surface water, and mixed sources) across the continental United States and its territories (Guelfo & Adamson, 2018). The UCMR3 database is currently the only publicly available drinking water database with routine nationwide PFAS monitoring data. However, studies utilizing this large dataset are relatively few (Andrews et al., 2021; Eaton et al., 2018; Guelfo & Adamson, 2018; Hu et al., 2016). Two previous studies focused exclusively on examining the UCMR3 PFAS data (Guelfo & Adamson, 2018; Hu et al., 2016). These studies, and others that have incorporated UCMR3 data into their analyses, have used varying aggregation techniques. Hu et al. (2016) aggregated analytical results at the eight-digit hydrologic unit code (HUC-8) level (U.S. EPA, 2013) and used the maximum concentrations detected in each watershed to formulate a regression model for predicting PFAS occurrence in drinking water (Hu et al., 2016). Guelfo and Adamson (2018) aggregated and analyzed data nationally by PWS characteristics (e.g., size and source water type) and temporally by quarterly sampling summary statistics (Guelfo & Adamson, 2018). Their work also examined all individual samples together (n = 36,972) to explore the possibility of statistical significance in co-contaminant mixtures (Guelfo & Adamson, 2018). In addition to these two focused analyses of UCMR3 data, Crone et al. (2019) provided UCMR3 results aggregated by water system size and water system source water and presented summary statistics for the frequency of occurrence of PFAS across all samples collected (Crone et al., 2019). Andrews and Naidenko (2020) included UCMR3 results as part of a broader assessment of population-wide exposure to PFAS (Andrews & Naidenko, 2020). To date, the work of Hu et al. (2016) is the first and only work to spatially analyze the UCMR3 data published in the scientific literature (Hu et al., 2016). Based on several broad assumptions, the authors estimated 16.5 million Americans potentially affected by at least one PFAS in drinking water supplies (Hu et al., 2016). Increasing exploitation of this and other large datasets provides opportunities for accelerating the current scientific understanding of PFAS in source and drinking water at a rate that allows stakeholders to effectively and expediently address risks associated with exposure.

Chapter 3. Development of a methodology to examine nationwide PFAS occurrence in drinking water

3.1 Introduction

Although the UCMR3 dataset is publicly available, beyond instructions for importing and filtering the data with Microsoft Excel (U.S. EPA, 2016a), no guidance has been issued to enable interested users to effectively use the data. Further, no study in the scientific literature has utilized the PWS and zip code information provided by the UCMR3 to spatially analyze national PFAS occurrence. In this chapter, the UCMR3 data are used to enhance the current understanding of nationwide PFAS occurrence in drinking water in the United States at the individual PWS-level. A straightforward data analysis methodology is presented for analyzing these data at multiple spatial scales (national, regional, and state) and visualizing PFAS occurrence at the finest spatial resolution possible. To the best of the author's knowledge, the results reported herein are the first to spatially analyze the results of the only currently existing nationwide database of PFAS in drinking water at the individual public water system (PWS) level. This methodology will allow stakeholders to identify geographic hotspots of PFAS occurrence; better anticipate possible results as monitoring efforts continue at lower quantitation and reporting levels; and initiate efficient data management and analysis frameworks. The methods presented can be applied to further exploit existing monitoring data and to rapidly generate meaningful insights as fifth UCMR (UCMR5) sampling round data are released, as early as mid-2023 and quarterly thereafter (U.S. EPA, 2022b).

3.2 Methods

3.2.1. Nationwide monitoring effort

The UCMR3 mandated all PWSs serving greater than 10,000 customers (and a representative sample of PWSs serving less than or equal to 10,000 customers) to participate in sampling. Altogether, 4,920 PWSs were required to collect and analyze samples for six PFAS, along with twenty-two other chemicals and two viruses, all of which were selected using a stepwise prioritization process considering possible adverse health effects, suspected occurrence, and the availability of a reliable analytical method (U.S. EPA, 2012a, 2012b). PWSs using surface water (SW) and/or groundwater under the direct influence of surface water were required to perform quarterly sampling at each EP (U.S. EPA, 2012b). In general, PWSs using only groundwater (GW) were required to collect two samples, five to seven months apart, from every EP (U.S. EPA, 2012b). However, large groundwater systems could submit a proposed sampling plan and, contingent on USEPA approval, conduct sampling at representative EPs rather than all EPs to their distribution network (U.S. EPA, 2012b). All samples collected for the UCMR3 were analyzed for six perfluoroalkyl substances, including three perfluoroalkylcarboxylic acids (PFCAs): perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), and perfluorononanoic acid (PFNA); and three perfluoroalkylsulfonic acids (PFSAs): perfluorobutanesulfonic acid (PFBS), perfluorohexanesulfonic acid (PFHxS), and perfluorooctane sulfonic acid (PFOS). Figure 3.1 illustrates the UCMR3 data structure for each PWS resulting from this monitoring regime, using a generic example of a PWS with one facility sourced by surface water; however, many PWSs have multiple facilities, which is discussed in greater detail in Chapter 4.

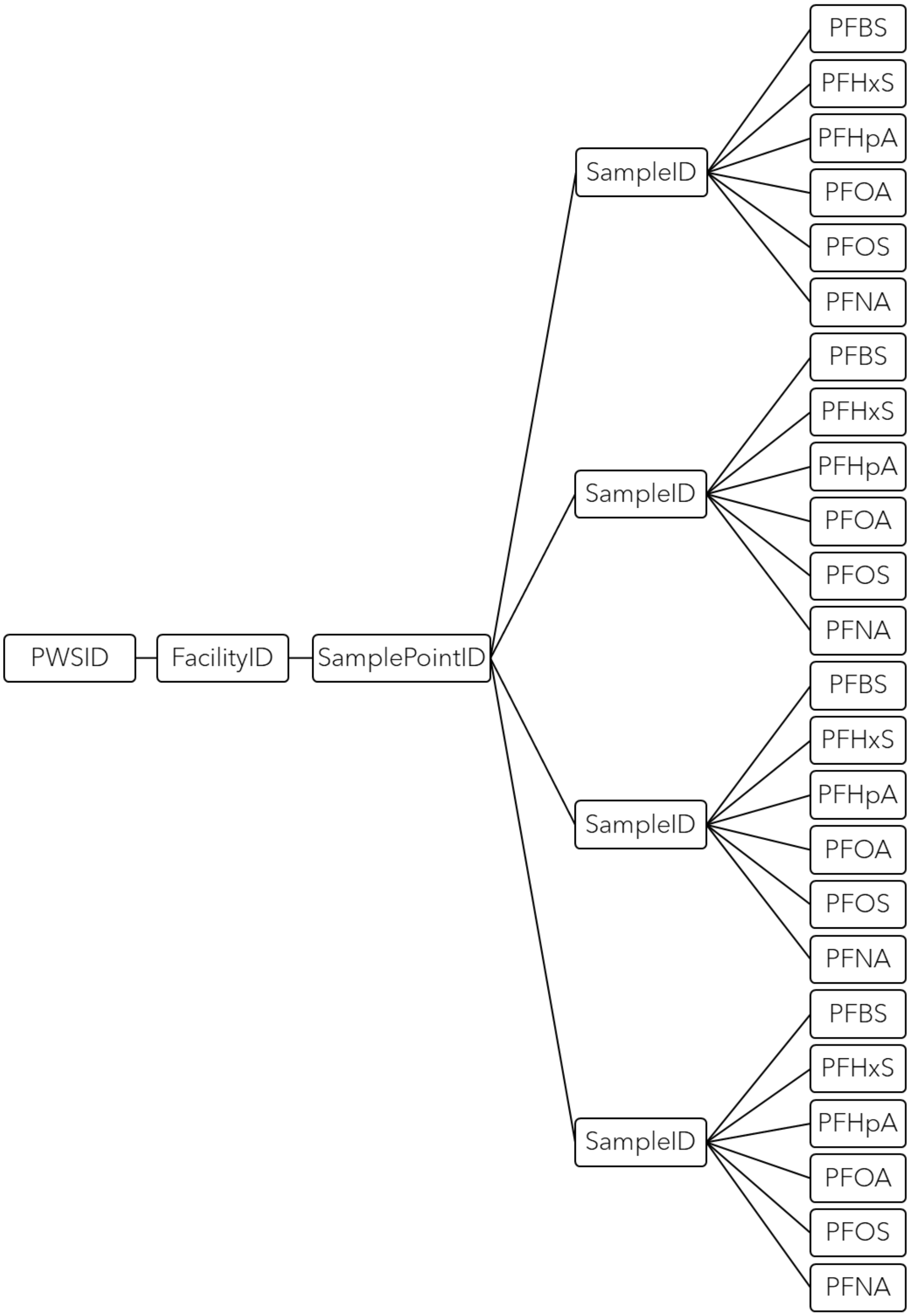


Figure 3.1. Example of UCMR3 data structure for a public water system (PWS) with one facility sourced by surface water.

Four quarterly sampling events were required at each facility sourced by surface water (SW), yielding 24 analytical results per facility. Two semi-annual sampling events were required at each facility sourced by groundwater (GW), yielding 12 analytical results per GW facility.

USEPA Method 537 was used to analyze all samples collected under UCMR3 (U.S. EPA, 2012b). As is routine for USEPA drinking water method validation (Boone et al., 2019), an interlaboratory comparison testing approach was used to establish minimum reporting levels (MRLs) for each contaminant (Table 3.1) to produce a quantitative dataset comparable across laboratories (U.S. EPA, 2012b).

Table 3.1. Perfluoroalkyl substances monitored during U.S. Environmental Protection Agency's (USEPA) Third Unregulated Contaminant Monitoring Rule (UCMR3).

Acronym	Chemical Name	Sub-Class[a]	MRL[b] (ng L^{-1})
PFBS	perfluorobutanesulfonic acid	PFSA	90
PFHxS	perfluorohexanesulfonic acid	PFSA	30
PFHpA	perfluoroheptanoic acid	PFCA	10
PFOA	perfluorooctanoic acid	PFCA	20
PFOS	perfluorooctanesulfonic acid	PFSA	40
PFNA	perfluorononanoic acid	PFCA	20

[a]perfluorosulfonic acid (PFSA) or perfluoroalkyl carboxylic acid (PFCA), [b]minimum reporting level

As previously discussed in the literature, MRLs established for the six PFAS included in the UCMR3 are higher than the method detection and quantitation levels of USEPA Method 537 (Boone et al., 2019; Eaton, 2017; Hu et al., 2016). The UCMR3 MRLs are also substantially higher than the USEPA's 2022 health advisory levels for PFOA and PFOS. The upcoming UCMR5 program, which will occur from January 2023 to December 2025, will quantify twenty-five PFAS using EPA Method 533 and four PFAS using EPA Method 537.1 (U.S. EPA, 2021e). These methods were published in 2019 and 2020, respectively, and will yield MRLs orders of magnitude lower than those of the UCMR3 program; however, given the extreme decrease in health advisory benchmarks for PFOA and PFOS in June 2022, UCMR5 MRLs will remain orders of magnitude greater than current health advisory levels (Table 3.2).

Table 3.2. USEPA UCMR minimum reporting levels (MRLs) and lifetime health advisory levels (LHAs).

PFAS	2012 UCMR3 MRL (ng L^{-1})	2016 USEPA LHA (ng L^{-1})	2021 UCMR5 MRL (ng L^{-1})	2022 USEPA LHA (ng L^{-1})
PFOA	20	70 (combined)	4	0.004
PFOS	40		4	0.02
GenX	N/A	N/A	5	20
PFBS	90	N/A	3	2000

3.2.2. Monitoring data

The UCMR3 ran officially from 2013-2015 (U.S. EPA, 2012b), with the large majority of analytical results obtained during this timeframe: 11,057 samples in 2013; 15,419 in 2014; and 10,102 in 2015. A small percentage of the total number of samples analyzed were collected in 2016 (394 samples, 1.1% of total). Over this time frame, a total of 36,972 drinking water samples were collected from distribution system entry points (EPs) at 4,920 PWSs across all American states and territories during the UCMR3 program. Resulting PFAS monitoring data can be accessed by downloading the UCMR3 data by method classification from the USEPA's occurrence data webpage and importing the "UCMR3_537" text file into Microsoft Excel (U.S. EPA, 2022c). With six PFAS results per sample, the UCMR3 dataset contains 221,831 PFAS concentration records in total, each of which include 21 additional attributes. Altogether, the UCMR3 PFAS dataset contains approximately 4.9 million data entries, which identify the PWS, treatment facility, distribution system entry point, and sampling event associated with each concentration record, along with additional information such as the facility's source water type and the date of sample collection. Names and definitions of primary data fields used for the analyses performed in this chapter are provided in Table 3.3 (U.S. EPA, 2017). All data field names and definitions are provided in Appendix Table A.1. U.S. postal zip codes for all areas served drinking water from each PWS monitored during UCMR3 are also available in a separate text file ("UCMR3_ZipCodes.txt") through the USEPA UCMR occurrence data webpage (U.S. EPA, 2017).

Table 3.3. Names and definitions of UCMR3 data fields used for spatial and temporal analyses (U.S. EPA, 2017)**.**

Field Name	**Definition**
PWSID	9-character Public Water System (PWS) identification code
CollectionDate	Date of sample collection (month, day, year)
SampleID	Identification code for each sample, as defined by the analytical laboratory
Contaminant	PFBS, PFHxS, PFHpA, PFOA, PFOS, or PFNA
AnalyticalResultsSign	"<" (less than MRL) or "=" (at or above the MRL)
AnalyticalResultValue	Analytical result in µg/L (null values represent concentrations < MRL)
Region	EPA region
State	State abbreviation

3.2.3. Data processing and aggregation

Microsoft Excel Tables and PivotTables were used to query, aggregate, and analyze the PFAS occurrence data at multiple spatial scales (national, regional, state, and individual PWS). The UCMR3 PFAS results include facility and EP identification identifiers, but do not include spatial location information for areas served by individual EPs. This is a design feature of the UCMR3 dataset: a draft version of UCMR3 required the zip codes of individual EP service areas to be reported. However, stakeholder concerns led to a final rule establishing that all zip codes served drinking water by the broad distribution network of each PWS would be provided in lieu of the zip codes served by the individual facilities and EPs within a PWS (U.S. EPA, 2012b). The consequences of this reporting decision on analyses of occurrence and assessments of population exposure risks are explored in detail in Chapter 4. The spatial analyses of PFAS occurrence at the PWS-level provided here represents the finest spatial resolution possible given the constraints of the UCMR3 dataset architecture.

Interpreting results of the following analyses of national PFAS occurrence requires a clear understanding of the approach used to aggregate the data at the individual PWS level. Importantly, minimum, maximum, and mean concentrations presented in the following sections neglect samples with PFAS levels below the UCMR3 MRLs. This approach follows from the limitations of the relatively high MRLs used in UCMR3, as previously discussed in the literature (Boone et al., 2019; Guelfo & Adamson, 2018; Hu et al., 2016). Accounting for results below MRLs by attributing a value of 0 ng L^{-1} would lead to a misinterpretation of UCMR data. For example, a small PWS with one distribution system EP sourced by groundwater exclusively would have collected two samples, 5-7 months apart, to fulfill the requirements of the UCMR3. Suppose the true concentrations of PFBS in these two samples are 110 and 80 ng L^{-1}. Since the MRL for PFBS was 90 ng L^{-1}, attributing a value of zero for records below MRLs would lead to an estimated average of 55 ng L^{-1}, which is 40 ng L^{-1} less than the true average. Furthermore, an analysis of groundwater monitoring results performed by Eaton et al. (2018) suggested that there is generally no significant difference in either concentrations or overall occurrence frequency of any PFAS between two sampling events at a given entry point (Eaton et al., 2018). Given the results of this 2018 study, a scenario in which a sample from a facility sourced by groundwater contains 110 ng L^{-1} PFBS and a sample collected 5-7 months later does not contain PFBS above quantitation levels is unlikely, unless specialized treatment strategies were implemented between the two sampling events. Consequently, an analysis that assigns a value of 0 ng L^{-1} for records below MRLs could potentially mask PFAS occurrence levels of concern for populations potentially served drinking water by a given PWS. These considerations will be important in understanding the pending UCMR5 data as well, since the UCMR5 MRL for both PFOA and PFOS will be 4 ng L^{-1}, while the 2022 USEPA health advisory levels are 0.004 ng L^{-1} (PFOA) and 0.02 ng L^{-1} (PFOS).

3.2.4. Spatial analysis

To visualize nationwide PFAS occurrence at the PWS-level resolution, the data were aggregated by taking the average of samples with PFAS levels exceeding UCMR3 MRLs. As discussed in the previous paragraph, samples with PFAS levels below the UCMR3 MRLs (i.e., samples with concentrations that were censored in the publicly available dataset) were neglected. Figure 3.2 builds from Figure 3.1 to clearly illustrate this aggregation method.

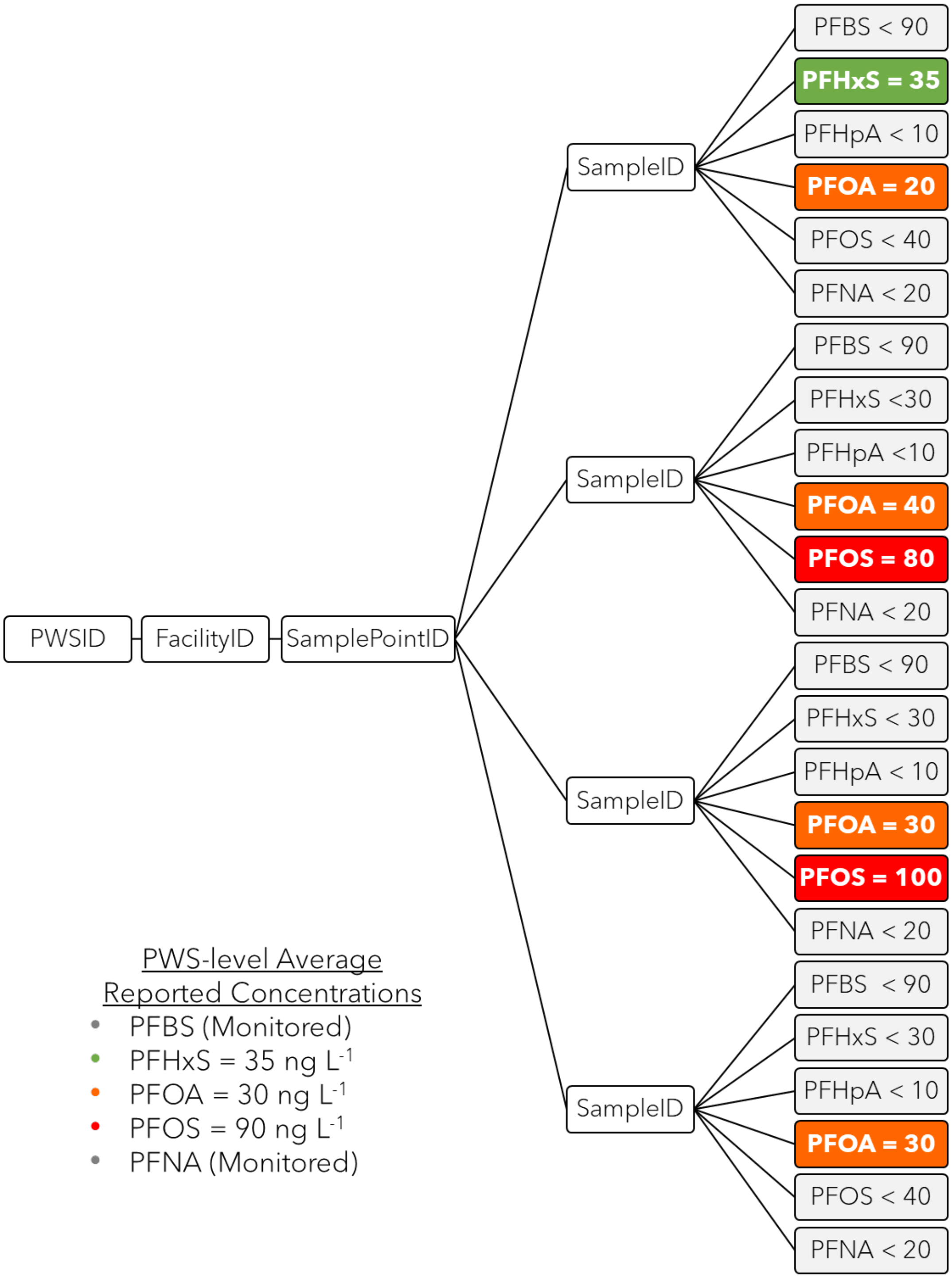


Figure 3.2. Illustration of PWS-level data aggregation method

As shown in this theoretical example, the number of analytical results used to determine average PWS-level concentrations of PFHxS, PFOA, and PFOS for visualization in the following occurrence maps would be 1, 4, and 2, respectively. Where no concentrations exceeded UCMR3 MRLs, the PWS was shown on the map to indicate that it was monitored.

The occurrence data follow a relational database model, so the PWS identification code (PWSID), which is common to both the PFAS occurrence dataset and zip code data table (U.S. EPA, 2017), was used as an identifier (Beaumont & Shadbolt, 2016) to link PFAS occurrence data to the zip codes of customers served drinking water by each of the PWSs. However, the USEPA-provided data table designating zip code(s) of areas served drinking water by each PWS (U.S. EPA, 2017) required validation and cleaning (Chu et al., 2016). For example, some zip codes were displayed with a hyphen (-), which prevented the zip codes from being matched to geographic locations. These inconsistencies were identified by filtering the zip code data table using Microsoft Excel's Tables feature and rectified using the Text to Columns tool (Data > Data Tools > Text to Columns). Additionally, zip codes for some PWSs were not provided. Missing zip codes were manually determined by searching online using additional data fields (e.g., state, PWSName, and FacilityName). After cleaning the data, Excel's XLOOKUP function was used to link the PFAS concentration data to the corresponding spatial information (zip codes). The resulting modified PFAS occurrence data table included an additional attribute, zip code, which was used to join PFAS occurrence data to spatial locations (the geographic coordinates of zip code centroids) via Esri ArcGIS 10.8. The zip code centroid layer package can be found through ArcGIS Online (Esri et al., 2021). Seventeen PWSs were not successfully matched to the zip code centroid layer because the zip codes provided in the occurrence data were not valid. These validation issues were also rectified manually (Appendix Table A.2). Once all validation issues were resolved, the PFAS occurrence data for the 4,920 PWSs monitored were spatially analyzed using Esri ArcGIS 10.8.

3.3 Results and Discussion

3.3.1. Nationwide PFAS occurrence

Table 3.4 provides the number of samples with concentrations exceeding MRLs for each PFAS monitored (out of the 36,972 total samples), and summary statistics of this subset of samples. For each PFAS, the state or territory where the maximum concentration was detected is provided. Where maximum concentrations occurred in an American territory, the maximum concentration detected within the continental United States and its associated state are also provided.

Table 3.4. Overview of UCMR3 samples with PFAS concentrations exceeding USEPA minimum reporting levels.

Analyte	n>MRL[a]	%>MRL[b]	Concentration (ng L^{-1})[c]			
			MRL	Min.	Max. (Location)[d]	Mean
PFBS	19	0.05	90	90	370 (GA)	184
PFHxS	207	0.56	30	30	1,600 (MP[e]) 730 (AZ)	139
PFHpA	236	0.64	10	10	410 (MP[e]) 87 (PA)	25
PFOA	379	1.03	20	20	349 (PA)	41
PFOS	292	0.79	40	40	7,000 (MP[e]) 1,800 (DE)	170
PFNA	19	0.05	20	22	56 (NJ)	36

[a] *Number of samples with PFAS concentrations exceeding minimum reporting level (MRL).* [b] *Percentage of samples exceeding MRL.* [c] *Summary statistics for reported concentrations of PFAS. Analytical records below MRLs (records with no numeric data) were excluded from determinations of minimum, maximum, and mean.* [d] *Where maximum concentrations occurred in U.S. territories, the maximum concentration within the continental U.S. and its associated state are also provided.* [e] *Northern Marianas Islands.*

As shown in Table 3.4, PFOA most frequently exceeded its MRL of 20 ng L^{-1}, despite its gradual phase-out beginning in 2006 (U.S. EPA, 2022b). The second most frequently reported PFAS was PFOS, despite having been voluntarily phased out of production in the U.S. by its primary manufacturer in 2002 (U.S. EPA, 2016b), more than a decade prior to UCMR3 monitoring. Given the current interim LHAs for PFOA and PFOS (0.004 and 0.02 ng L^{-1}, respectively), the concentrations of PFOA in each of the 379 samples reported in the UCMR3 data were, at minimum, 5,000 times greater than the updated interim health advisory. The 292 samples exceeding the UCMR3 MRL for PFOS contained concentrations at least 2,000 times the new advisory level. In reviewing these and the following results, it is important to acknowledge data gaps due to MRLs that are much higher than the threshold concentrations of concern to human health based on the USEPA's most recent assessments. Notably, one of several commercial laboratories responsible for analyzing approximately 10,500 (~30%) of the UCMR3 samples for PFAS, Eurofins Eaton Analytical, reported occurrence frequencies using MRLs much lower than the UCMR3 MRL (5 and 2.5 ng L^{-1}) (Eaton, 2017). Using a 5 ng L^{-1} reporting level, occurrence frequencies for their subset of UCMR3 samples (which included samples from all states and territories) for PFOA, PFOS, and PFBS were 12.5%, 11.5%, and 5.3%, respectively. At the 2.5 ng L^{-1} reporting level, these frequencies increased to 23.5%, 20.5%, and 11.9% for PFOA, PFOS, and PFBS, respectively. Even higher national frequencies of detection (76 to 96%) were observed in a nationwide study of twenty-five drinking water treatment plants that analyzed samples in a single laboratory and used the USEPA lowest concentration minimal reporting level (LCMRL) process to set MRLs for the PFAS monitored during UCMR 3 ranging from 0.03 to 0.56 ng L^{-1} (Boone et al., 2019; Eaton, 2017). These findings may help stakeholders anticipate UCMR5 results, given that the upcoming UCMR5 data will use an MRL of 4 ng L^{-1} for PFOA and PFOS, and 3 ng L^{-1} for PFBS.

3.3.2. Regional, state, and PWS-level occurrence

All ten EPA regions reported at least one PWS with a PFAS detection exceeding UCMR3 MRLs (Appendix Table A.3). Of the 10 EPA regions (Figure 3.3), the maximum number of analytical results above MRLs, 239 (0.6% of regional results), occurred in EPA Region 4, and the highest frequency of detections above MRLs occurred in Region 8 (208 analytical results, 2.2% of total). Of the records exceeding MRLs in Region 8, 206 (99.0%) are from PWSs in Colorado, where the state-level frequency of MRL exceedance was 6.1%, the highest among the U.S. states. However, the Northern Marianas Islands surpassed this frequency, with 21 detections above MRLs (7.8% of analytical results). Regional and state-level occurrence findings for PFOA and PFOS, the two legacy PFAS which have historically been the subject of the most research and regulatory attention, are presented in Tables 3.5 and 3.6, respectively. Each of these tables are paired with a corresponding PWS-level spatial analysis of occurrence in the continental United States, Figure 3.4 and Figure 3.5. These spatial analyses reveal geographic regions with elevated levels of these two legacy PFAS, such as the Tennessee Valley region of North Alabama, the Los Angeles and Orange County areas of Southern California, and much of the Northeast. Additionally, the analyses identify several isolated areas with PFOA and PFOS levels of concern.

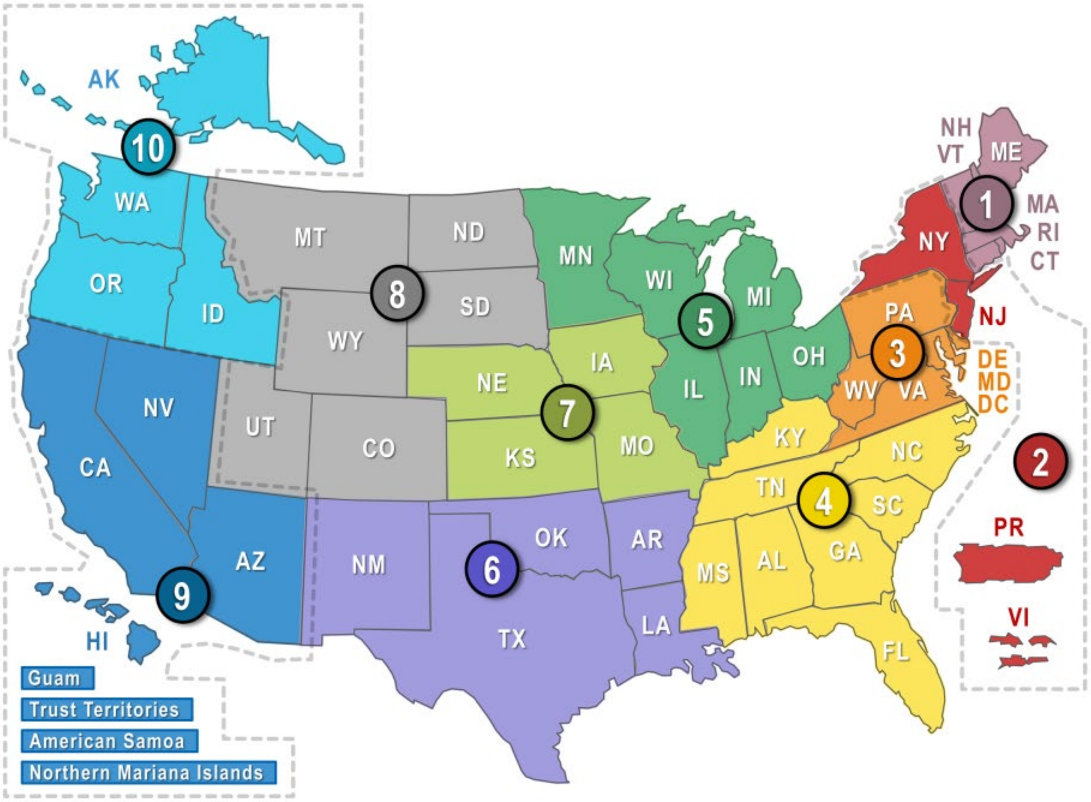


Figure 3.3. EPA Region Map

Source: United States Environmental Protection Agency (U.S. EPA, 2022a)

Table 3.5. Regional and state-level summary of UCMR3 samples with PFOA concentrations exceeding minimum reporting level (20 ng L^{-1}) and 2022 USEPA health advisory level, 0.004 ng L^{-1}.

EPA Region	**Samples Analyzed, n**	**n>MRL & 2022 LHA**	**%n>MRL & 2022 LHA**	**Concentration (ng L^{-1})[a]**		
State				**Min.**	**Max.**	**Mean**
Region 1	**1933**	**18**	**0.93**	**20**	**81**	**35**
MA	1208	10	0.83	20	62	33
NH	125	5	4.00	20	67	35
RI	112	3	2.68	20	81	42
Region 2	**3702**	**99**	**2.67**	**20**	**110**	**30**
NJ	1461	87	5.95	20	110	30
NY	1684	12	0.71	22	48	31
Region 3	**2371**	**51**	**2.15**	**20**	**349**	**72**
DE	130	14	10.77	21	140	52
MD	227	1	0.44	-	-	21
PA	1360	27	1.99	20	349	82
VA	502	1	0.2	-	-	22
WV	136	8	5.88	41	129	84
Region 4	**6937**	**67**	**0.97**	**20**	**100**	**31**
AL	1056	32	3.03	20	100	33
FL	1460	6	0.41	21	65	34
GA	755	14	1.85	20	70	34
KY	617	2	0.32	-	-	20
NC	1320	11	0.83	20	31	24
SC	463	1	0.22	-	-	24
TN	652	1	0.15	-	-	20
Region 5	**4211**	**20**	**0.47**	**20**	**338**	**71**
IL	1288	3	0.23	23	59	41
MN	444	11	2.48	28	338	103
OH	663	3	0.45	20	27	24
WI	651	3	0.46	22	43	30
Region 6	**4656**	**3**	**0.06**	**26**	**40**	**33**
OK	513	2	0.39	34	40	37
TX	2958	1	0.03	-	-	26
Region 8	**1545**	**52**	**3.37**	**20**	**90**	**47**
CO	565	52	9.2	20	90	47
Region 9	**8501**	**63**	**0.74**	**20**	**220**	**36**
AZ	1108	15	1.35	20	50	38
CA	6468	46	0.71	20	53	28
MP[b]	45	2	4.44	200	220	210
Region 10	**1881**	**6**	**0.32**	**20**	**51**	**29**
WA	1039	6	0.58	20	51	29

[a] *Analytical records below MRLs (records with no numeric data) were excluded from determinations of minimum, maximum, and mean.* [b] *Northern Marianas Islands.*

All states within USEPA Region 3 reported at least one sample containing PFOA at levels exceeding the UCMR3 MRL of 20 ng L^{-1}. PFOA was reported in 10.8% of the 130 samples collected in Delaware, the highest occurrence frequency among all U.S. states and territories. The maximum concentration of PFOA in any sample across the U.S., 349 ng L^{-1}, was collected from Pennsylvania, where 2.0% of the 1,360 analyzed samples exceeded the PFOA MRL. This concentration exceeded the maximum concentration reported in Delaware by a factor of 2.5. The overall geographic distribution of PFOA occurrence in the continental United States is provided in Figure 3.4.

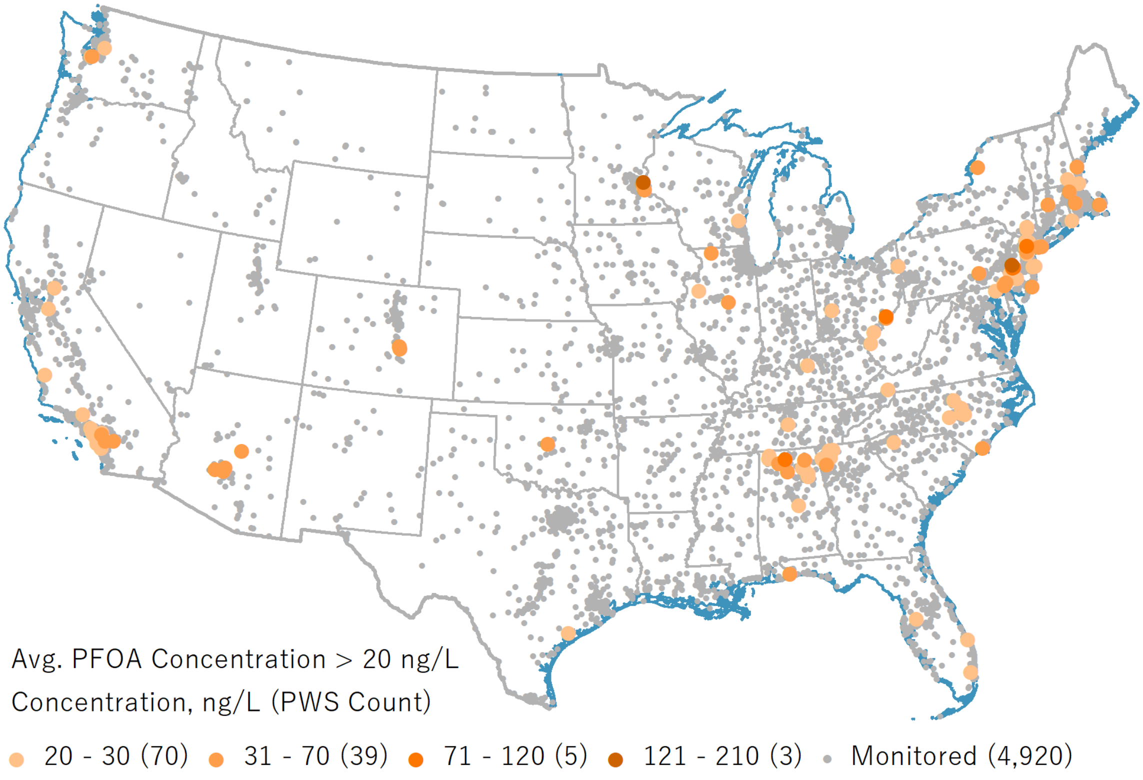


Figure 3.4. PWS-level spatial analysis of average reported concentrations of PFOA during UCMR3.

Points represent centroids of zip codes served by PWSs.

The USEPA region with the greatest PFOA occurrence frequency (3.4%) was Region 8, which covers six states: Colorado, Montana, North Dakota, South Dakota, Utah, and Wyoming. Notably, all PFOA concentrations reported above UCMR3 MRLs can be attributed to just one state, Colorado. Moreover, examining the occurrence data at the PWS-level reveals that all samples with reported concentrations of PFOA were collected from very few PWSs (Figure 3.4). In fact, all PFOA MRL exceedances can be attributed to just three PWSs, two of which provide drinking water to the same zip code in southeastern Colorado Springs (80911), and the other to an adjacent zip code just outside of the Colorado Springs city limit (80817). PFOA occurrence in the southeastern U.S. (Region 4), was much more widespread, with at least one sample exceeding the PFOA MRL in all Region 4 states except Mississippi (Table 3.5, Figure 3.4). In Region 4, PFOA occurrence was especially prevalent at PWSs in Northwest Georgia and North Alabama.

The regional occurrence frequency of PFOS (Table 3.5, Table 3.6) was less than PFOA across 8 of the 10 USEPA regions (all except Regions 5 and 9). This observation could be attributed to the difference between reporting levels, since the UCMR3 MRL for PFOS was 40 ng L^{-1}, whereas the MRL for PFOA was 20 ng L^{-1}. However, in every EPA region, the maximum reported concentrations of PFOS exceeded that of PFOA. In EPA Regions 1, 2, and 3, maximum concentrations of PFOS were approximately 5 times greater than PFOA. The most substantial difference was observed in Region 9, where the maximum concentrations of both PFOA and PFOS were detected in the Northern Marianas Islands. The maximum concentration of PFOS (7,000 ng L^{-1}) exceeded that of PFOA (220 ng L^{-1}) by a factor of nearly 32. While PFOA was not reported in any sample in the Salt River Pima-Maricopa Indian Community, the American Samoa, or Guam, PFOS was reported in at least one sample from each of these territories. The PWS-level spatial analysis of PFOS occurrence in the continental United States is provided in Figure 3.5.

Table 3.6. Regional and state-level summary of UCMR3 samples with PFOS concentrations exceeding minimum reporting level (40 ng L^{-1}) and 2022 USEPA health advisory level, 0.02 ng L^{-1}.

EPA Region	**Samples**	**n>MRL &**	**%n>MRL**	**Concentration (ng L^{-1})[a]**		
State	**Analyzed, n**	**2022 LHA**	**& 2022 LHA**	**Min.**	**Max.**	**Mean**
Region 1	**1933**	**15**	**0.78**	**50**	**430**	**160**
MA	1208	11	0.91	60	430	154
ME	82	3	3.66	50	290	197
NH	125	1	0.8	-	-	120
Region 2	**3702**	**25**	**0.68**	**41**	**530**	**114**
NJ	1461	12	0.82	43	98	54
NY	1684	13	0.77	41	530	170
Region 3	**2371**	**28**	**1.18**	**41**	**1800**	**333**
DE	130	4	3.08	130	1800	760
PA	1360	21	1.54	41	1090	288
WV	136	3	2.21	74	86	80
Region 4	**6937**	**62**	**0.89**	**40**	**380**	**76**
AL	1056	23	2.18	40	180	61
FL	1460	21	1.44	40	380	102
GA	755	8	1.06	45	120	74
KY	617	2	0.32	50	59	54
NC	1320	8	0.61	41	90	61
Region 5	**4211**	**22**	**0.52**	**43**	**439**	**142**
IL	1288	1	0.08	-	-	180
IN	437	1	0.23	-	-	78
MI	716	3	0.42	43	60	51
MN	444	7	1.58	46	439	183
OH	663	8	1.21	45	400	155
WI	651	2	0.31	53	140	97
Region 6	**4656**	**2**	**0.04**	**42**	**46**	**44**
TX	2958	2	0.07	42	46	44
Region 8	**1545**	**45**	**2.91**	**40**	**1300**	**161**
CO	565	45	7.96	40	1300	161
Region 9	**8501**	**91**	**1.07**	**40**	**7000**	**207**
09[b]	59	1	1.69	-	-	120
AS[c]	68	1	1.47	-	-	40
AZ	1108	14	1.26	44	300	116
CA	6468	60	0.93	41	156	57
GU[d]	206	8	3.88	41	410	145
MP[e]	45	7	15.56	42	7000	1780

EPA Region	**Samples**	**n>MRL &**	**%n>MRL**	**Concentration (ng L^{-1})^a**		
State	**Analyzed, n**	**2022 LHA**	**& 2022 LHA**	**Min.**	**Max.**	**Mean**
Region 10	**1881**	**2**	**0.11**	**514**	**600**	**557**
WA	1039	2	0.19	514	600	557

[a] *Analytical records below MRLs (records with no numeric data) were excluded from determinations of minimum, maximum, and mean.* [b] *Salt River Pima-Maricopa Indian Community.* [c] *American Samoa.* [d] *Guam.* [e] *Northern Marianas Islands.*

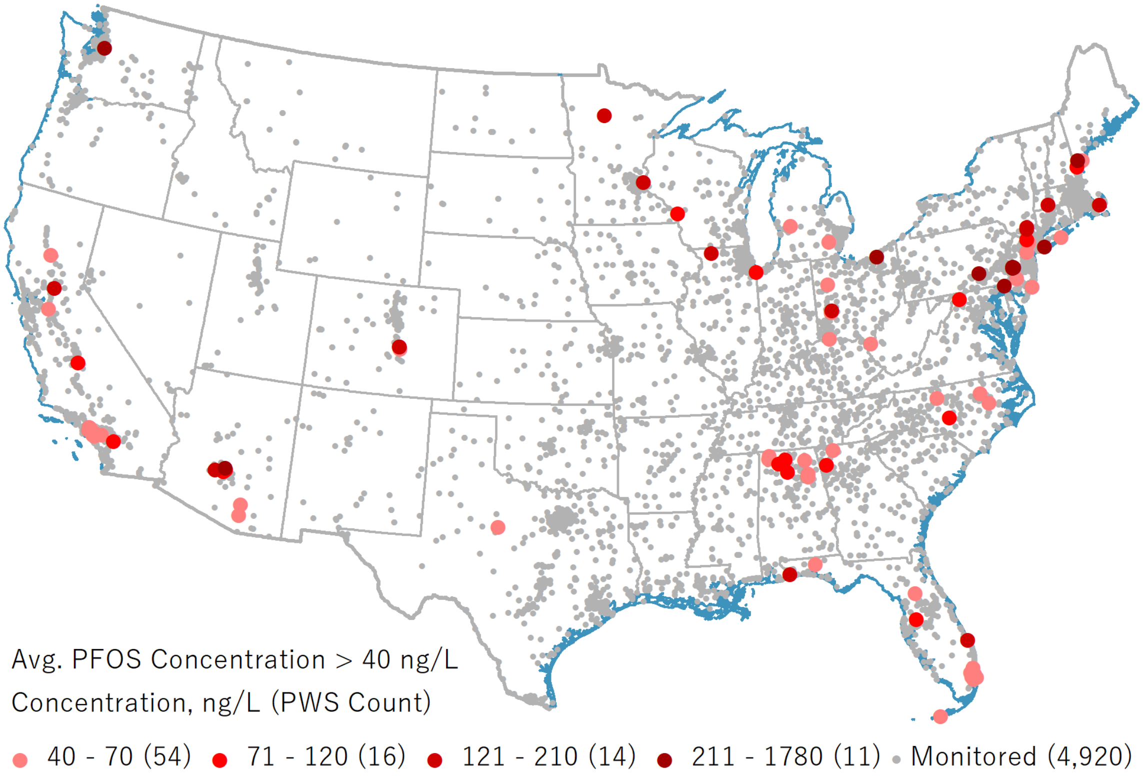


Figure 3.5. PWS-level spatial analysis of average reported concentrations of PFOS during UCMR3.

Points represent centroids of zip codes served by PWSs.

The geographic distribution of PFOS occurrence closely follows that of PFOA. In total, three more states reported PFOA in at least one sample than PFOS within the continental U.S. (23 compared to 26), but again, this could be due to the discrepancy in MRLs between the two compounds. In most states where PFOA was reported, PFOS was reported as well, though not necessarily in the same areas. The states where PFOA was reported, but no sample exceeded PFOS reporting levels are Rhode Island, Maryland, Virginia, South Carolina, Tennessee, and Oklahoma. In Maine, Indiana, and Michigan, PFOS concentrations were reported but no PFOA concentrations were reported. PFOA, PFOS, and PFBS are the three PFAS included in the UCMR3 for which individual health advisories have been issued by the USEPA as of June 2022. Analyses of regional, state-level, and PWS-level PFBS occurrence are provided in Table 3.7 and Figure 3.6.

Table 3.7. Regional and state-level summary of UCMR3 samples with PFBS concentrations exceeding minimum reporting level, 90 ng L^{-1} (2022 USEPA health advisory level = 2,000 ng L^{-1}).

EPA Region	**Samples Analyzed, n**	**n>MRL**	**%n>MRL**	**Concentration (ng L^{-1})**[a]		
State				**Min.**	**Max.**	**Mean**
Region 1	**1933**	**0**	**0.00**	**-**	**-**	**-**
Region 2	**3702**	**0**	**0.00**	**-**	**-**	**-**
Region 3	**2371**	**1**	**0.04**	**110**	**110**	**110**
PA	1360	1	0.07	110	110	110
Region 4	**6937**	**12**	**0.17**	**100**	**370**	**211.7**
AL	1056	3	0.28	100	150	126.7
GA	755	9	1.19	120	370	240
Region 5	**4211**	**0**	**0.00**	**-**	**-**	**-**
Region 6	**4656**	**0**	**0.00**	**-**	**-**	**-**
Region 7	**1235**	**0**	**0.00**	**-**	**-**	**-**
Region 8	**1545**	**4**	**0.26**	**90**	**150**	**110**
CO	565	4	0.71	90	150	110
Region 9	**8501**	**2**	**0.02**	**180**	**220**	**200**
MP[b]	45	2	4.44	180	220	200
Region 10	**1881**	**0**	**0.00**	**-**	**-**	**-**

[a] *Analytical records below MRLs (records with no numeric data) were excluded from determinations of minimum, maximum, and mean.* [b] *Northern Marianas Islands.*

Nineteen samples across four states and one territory exceeded the MRL for PFBS during the UCMR3 (Table 3.7). The MRL for PFBS was the highest among the six PFAS analyzed during the program: 90 ng L^{-1}. Nonetheless, the USEPA LHA for PFBS published in 2022 is 2,000 ng L^{-1}, and the maximum concentration detected nationwide during UCMR3 was 370 ng L^{-1}.

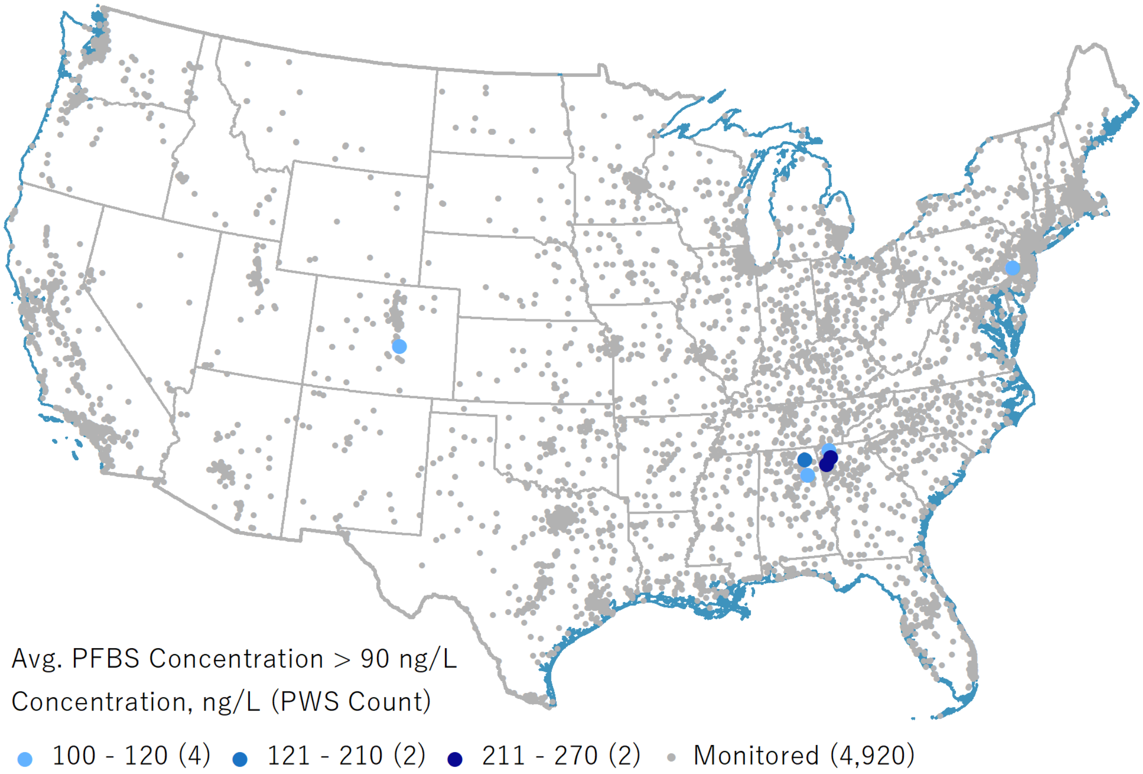


Figure 3.6. PWS-level spatial analysis of average reported concentrations of PFBS during UCMR3.

Points represent centroids of zip codes served by PWSs.

Although the nationwide distribution of PFBS was significantly less than PFOA and PFOS (likely due to the high PFBS MRL), the spatial analysis in Figure 3.6 reveals that geographic PFBS occurrence correlates with areas of substantial PFOA and PFOS occurrence: Colorado Springs, northeast Alabama, northwest Georgia, and southeast Pennsylvania.

The regional and state-level occurrence of PFHpA is provided in Table 3.8, followed by its national spatial distribution in Figure 3.7. The MRL of PFHpA (10 ng L^{-1})

was half of the PFOA MRL and a quarter of the PFOS MRL, but the overall occurrence frequency was less than both (Table 3.8).

Table 3.8. Regional and state-level summary of UCMR3 samples with PFHpA concentrations exceeding minimum reporting level, 10 ng L^{-1}.

EPA Region	**Samples Analyzed, n**	**n>MRL**	**%n>MRL**	**Concentration (ng L^{-1})^a**		
State				**Min.**	**Max.**	**Mean**
Region 1	**1933**	**16**	**0.83**	**10**	**58**	**18**
MA	1208	14	1.16	10	58	19
NH	125	2	1.6	12	15	14
Region 2	**3702**	**20**	**0.54**	**10**	**82**	**22**
NJ	1461	7	0.48	10	17	12
NY	1684	12	0.71	14	82	29
PR	547	1	0.18	10	10	10
Region 3	**2371**	**34**	**1.43**	**10**	**87**	**29**
DE	130	15	11.54	10	80	27
PA	1360	16	1.18	11	87	33
VA	502	1	0.2	12	12	12
WV	136	2	1.47	14	15	15
Region 4	**6937**	**80**	**1.15**	**10**	**69**	**20**
AL	1056	31	2.94	10	30	14
FL	1460	10	0.68	11	69	24
GA	755	8	1.06	10	40	20
NC	1320	29	2.2	10	60	24
SC	463	1	0.22	12	12	12
TN	652	1	0.15	11	11	11
Region 5	**4211**	**7**	**0.17**	**11**	**53**	**24**
MN	444	6	1.35	11	53	26
OH	663	1	0.15	11	11	11
Region 6	**4656**	**3**	**0.06**	**12**	**16**	**14**
NM	337	1	0.3	12	12	12
TX	2958	2	0.07	15	16	16
Region 7	**1235**	**-**	**-**	**-**	**-**	**-**
Region 8	**1545**	**48**	**3.11**	**10**	**70**	**26**
CO	565	48	8.5	10	70	26
Region 9	**8501**	**25**	**0.29**	**10**	**410**	**42**
09[a]	59	1	1.69	11	11	11

EPA Region	**Samples Analyzed,**	**n>MRL**	**%n>MRL**	**Concentration (ng L^{-1})^{a}**		
State	**n**			**Min.**	**Max.**	**Mean**
AZ	1108	6	0.54	11	30	19
CA	6468	12	0.19	10	22	14
GU	206	1	0.49	14	14	14
MP	45	5	11.11	16	410	150
Region 10	**1881**	**3**	**0.16**	**13**	**26**	**21**
WA	1039	3	0.29	13	26	21

[a] *Analytical records below MRLs (records with no numeric data) were excluded from determinations of minimum, maximum, and mean.*

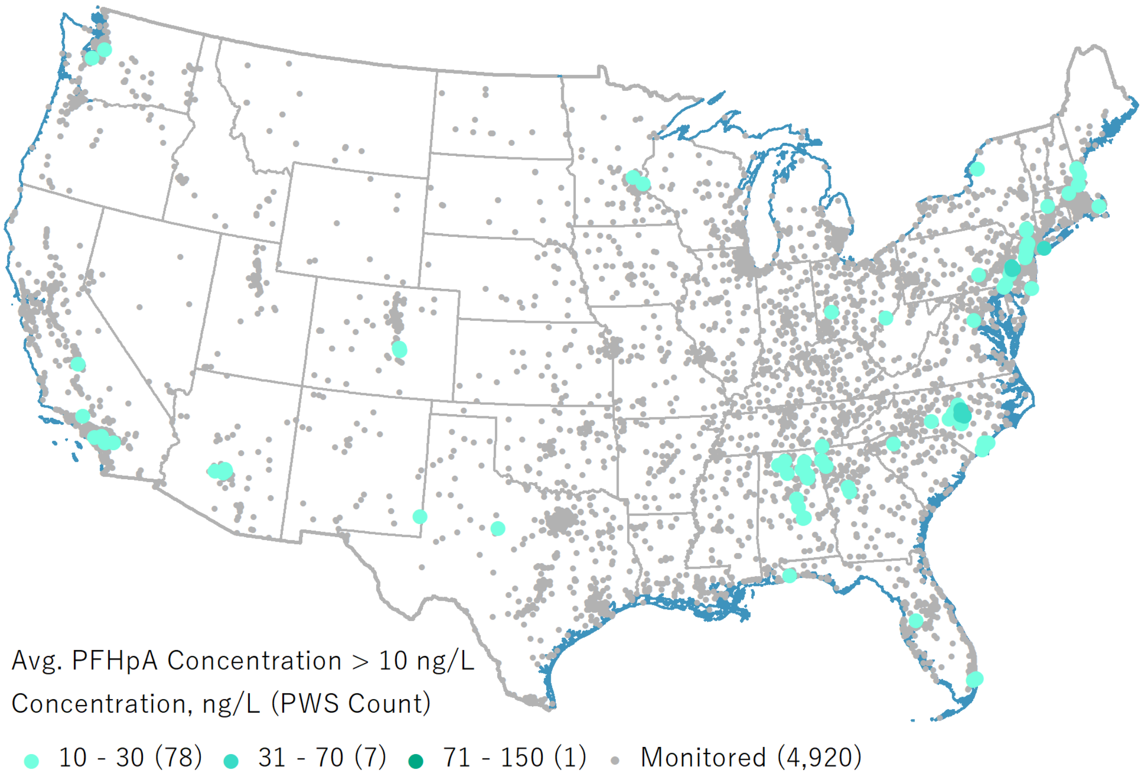


Figure 3.7. PWS-level spatial analysis of average reported concentrations of PFHpA during UCMR3.

Points represent centroids of zip codes served by PWSs.

The average reported concentrations of PFHpA at each PWS were much lower than PFOA, PFOS, and PFBS. The average reported PFHpA concentration exceeded 30 ng L^{-1}

at only 8 PWSs (Figure 3.7), whereas average reported concentrations of PFOA, PFOS, and PFBS exceeded 30 ng L^{-1}, at 47, 95, and 8 PWSs, respectively (Figures 3.3, 3.4, and 3.5), and due to MRL limitations, PWSs with average PFBS concentrations between 30 ng L^{-1} and 90 ng L^{-1} are not accounted for in these totals. PFHpA occurrence was abundant in North Carolina, and Alabama, and the Northeast, areas where many clustered PWSs reported concentrations of other PFAS, but less abundant where more isolated PWSs reported PFAS occurrence, like the Midwest and northern California. Interestingly, PFHpA was the only PFAS detected in New Mexico for the duration of the UCMR, near New Mexico's eastern border.

The regional and state-level occurrence of PFHxS is provided in Table 3.9, followed by its national spatial distribution in Figure 3.8.

Table 3.9. Regional and state-level summary of UCMR3 samples with PFHxS concentrations exceeding minimum reporting level, 30 ng L^{-1}.

EPA Region	**Samples Analyzed, n**	**n>MRL**	**%n>MRL**	**Concentration (ng L^{-1})^{a}**		
State				**Min.**	**Max.**	**Mean**
Region 1	**1933**	**13**	**0.67**	**32**	**270**	**88**
MA	1208	11	0.91	32	270	88
ME	82	2	2.44	70	110	90
Region 2	**3702**	**22**	**0.59**	**43**	**140**	**69**
NJ	1461	9	0.62	49	94	65
NY	1684	13	0.77	43	140	71
Region 3	**2371**	**29**	**1.22**	**31**	**680**	**155**
DE	130	4	3.08	50	680	315
PA	1360	20	1.47	31	590	146
WV	136	5	3.68	60	71	66
Region 4	**6936**	**17**	**0.25**	**34**	**180**	**79**
FL	1460	12	0.82	45	180	86
NC	1320	5	0.38	34	110	64
Region 5	**4211**	**17**	**0.4**	**32**	**187**	**85**
IL	1288	1	0.08	-	-	32

EPA Region State	**Samples Analyzed, n**	**n>MRL**	**%n>MRL**	**Concentration (ng L^{-1})[a]**		
				Min.	**Max.**	**Mean**
IN	437	1	0.23	-	-	65
MN	444	7	1.58	36	187	68
OH	663	5	0.75	39	150	123
WI	651	3	0.46	32	117	83
Region 6	**4656**	**4**	**0.09**	**47**	**63**	**52**
OK	513	2	0.39	48	63	56
TX	2958	2	0.07	47	52	49
Region 7	**1235**	**1**	**0.08**	**-**	**-**	**37**
KS	216	1	0.46	37	37	37
Region 8	**1545**	**59**	**3.82**	**30**	**590**	**191**
CO	565	57	10.09	30	590	197
SD	107	2	1.87	40	53	47
Region 9	**8501**	**43**	**0.51**	**30**	**1600**	**159**
09[a]	59	1	1.69	-	-	46
AZ	1108	12	1.08	30	730	218
CA	6468	15	0.23	32	120	60
GU[b]	206	10	4.85	30	140	57
MP[c]	45	5	11.11	65	1600	539
Region 10	**1881**	**2**	**0.11**	**201**	**241**	**221**
WA	1039	2	0.19	201	241	221

[a] Analytical records below MRLs (records with no numeric data) were excluded from determinations of minimum, maximum, and mean.

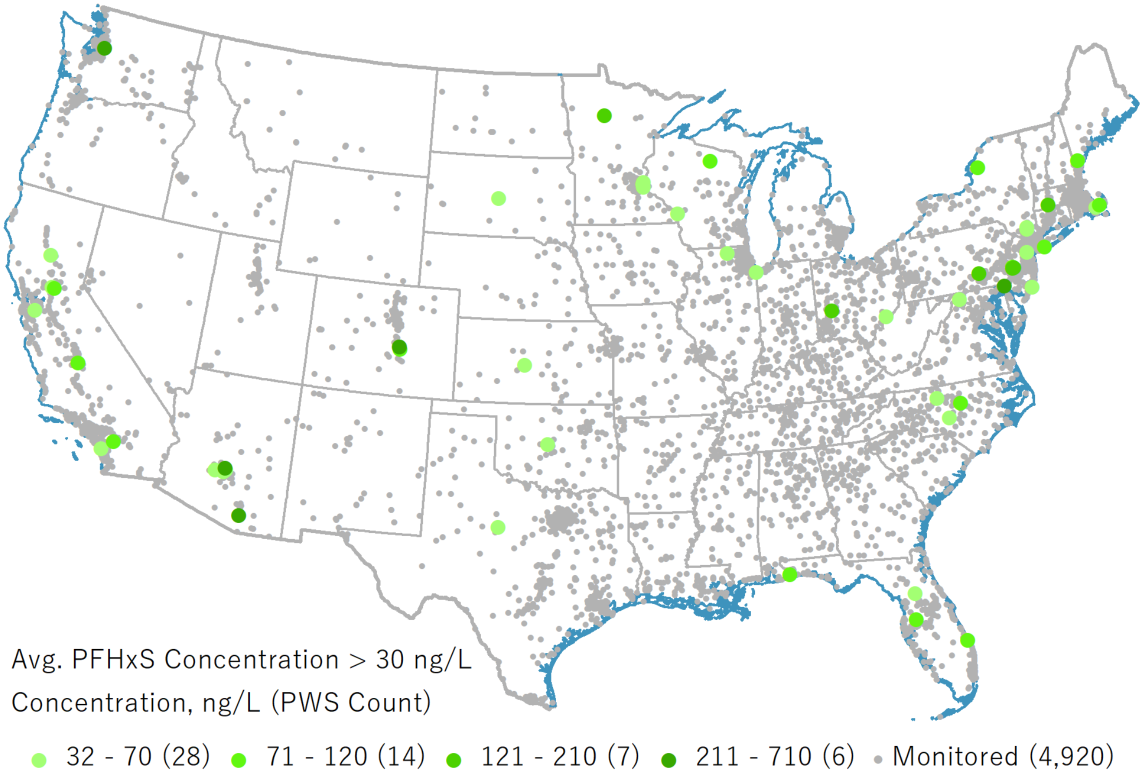


Figure 3.8. PWS-level spatial analysis of average reported concentrations of PFHxS during UCMR3.

Points represent centroids of zip codes served by PWSs.

Overall, PFHxS was reported at 55 PWSs (Figure 3.8). Clustering of PFOA, PFOS, PFBS, and PFHpA were observed in North Alabama and northwest Georgia; however, no samples exceeded the PFHxS MRL of 30 ng L^{-1} in this region. On the other hand, PFHxS was the only of the six PFAS reported in South Dakota and Kansas for the entire UCMR3 duration.

The regional and state-level occurrence of PFNA is provided in Table 3.10, followed by its national spatial distribution in Figure 3.9.

Table 3.10. Regional and state-level summary of UCMR3 samples with PFNA concentrations exceeding minimum reporting level, 20 ng L^{-1}.

EPA Region State	**Samples Analyzed, n**	**n>MRL**	**%n>MRL**	**Concentration (ng L^{-1})^{a}**		
				Min.	**Max.**	**Mean**
Region 1	**1933**	**1**	**0.05**	**-**	**-**	**35**
MA	1208	1	0.08	-	-	35
Region 2	**3702**	**8**	**0.22**	**24**	**56**	**40**
NJ	1461	7	0.48	24	56	41
NY	1684	1	0.06	32	32	32
Region 3	**2371**	**7**	**0.3**	**22**	**54**	**35**
DE	130	1	0.77	54	54	54
PA	1360	6	0.44	22	47	32
Region 4	**6937**	**1**	**0.01**	**-**	**-**	**27**
FL	1460	1	0.07	-	-	27
Region 5	**4211**	**0**	**0.00**	**-**	**-**	**-**
Region 6	**4656**	**0**	**0.00**	**-**	**-**	**-**
Region 7	**1235**	**0**	**0.00**	**-**	**-**	**-**
Region 8	**1545**	**0**	**0.00**	**-**	**-**	**-**
Region 9	**8501**	**0**	**0.00**	**-**	**-**	**-**
Region 10	**1881**	**2**	**0.11**	**27**	**28**	**28**
WA	1039	2	0.19	27	28	28

[a] *Analytical records below MRLs (records with no numeric data) were excluded from determinations of minimum, maximum, and mean.*

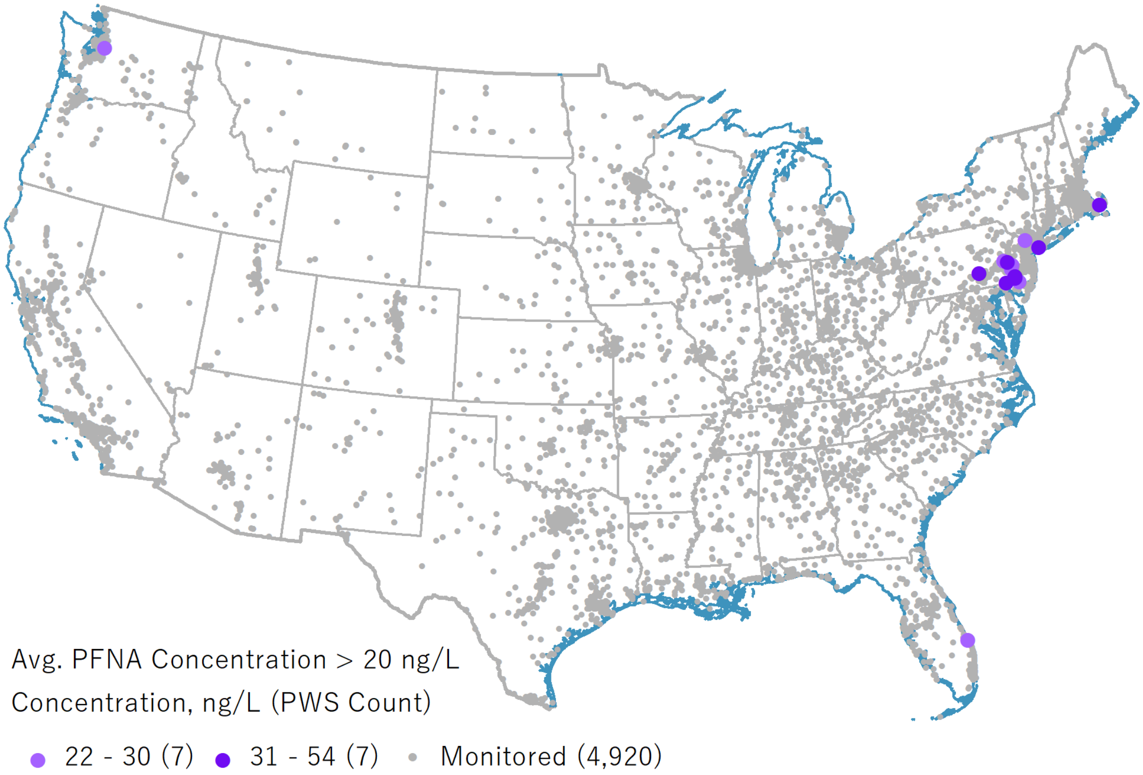


Figure 3.9. PWS-level spatial analysis of average reported concentrations of PFNA during UCMR3.

Points represent centroids of zip codes served by PWSs.

Unlike PFBS, the low occurrence of PFNA across the U.S. cannot be attributed to its MRL, 20 ng L^{-1}, which was lower than that of all PFAS monitored except PFHpA. All PFNA detections were in areas where legacy long-chain PFOA and PFOS had been reported as well. PFNA is the only PFAS monitored during the UCMR3 with no concentrations reported outside the continental U.S.

3.4 Conclusions

Using widely accessible software, Microsoft Excel and Esri ArcGIS, the work detailed in this chapter was effective in developing an efficient, accessible data analysis framework to further the utility of costly PFAS monitoring data. This work furthers the current understanding of the magnitude and extent of PFAS occurrence in drinking water by spatially analyzing the most expansive drinking water monitoring data to date at the finest resolution possible, the PWS-level. The approach presented here can be applied to rapidly generate meaningful spatiotemporal insights as fifth UCMR sampling round data are released, as early as mid-2023 (U.S. EPA, 2022b). The fine resolution of the spatial analyses presented allows for the identification and analysis of areas and individual PWSs where PFAS contamination is of particular concern, which can help utilities adjacent to those where PFAS were reported better understand the risks of PFAS contamination in their source water. This is especially useful for small utilities that were not required to monitor during UCMR3. The work creates a foundation for evaluating exposure risks as the current understanding of PFAS health effects and regulatory climate continue to evolve and identifying study sites of interest for further study (Chapter 4).

Because previous studies have indicated no statistically significant difference between PFAS concentrations in source water and drinking water in systems using traditional treatment methods, spatial analyses of available large datasets can assist state agencies in identifying source waters with the greatest risk of PFAS contamination. Moreover, utilities with limited resources, especially those with no available PFAS monitoring data to date (Guelfo & Adamson, 2018; Hu et al., 2016; Sunderland et al., 2018), can utilize spatial analyses to better anticipate risks of PFAS contamination in their utilities' source water. For example, small surface water-sourced facilities can gain insights from the PFAS monitoring results at systems upstream. Existing PFAS data from groundwater facilities can be used to better understand contamination risks at utilities using the same aquifer. Further, initiating data management and analysis frameworks with currently available data will expedite analyses of pending data, such as the UCMR5 database.

Further, “PFAS move through the environment in endless cycles between engineered (e.g., landfills, water and wastewater treatment facilities, etc.) and natural systems. With regulations that focus on managing PFAS within already engineered systems, where PFAS tend to be relatively concentrated or can more readily be concentrated and destroyed, we will be able to more cost-effectively reduce widespread contamination and exposures over the long term” (Crimi et al., 2022). As resources are allocated to the destruction of PFAS in these engineered systems, maximizing the utilization of nationwide data can aid in identifying where PFAS are most concentrated, and therefore foster a greater return on investment as sites are selected for the installation of advanced treatment and destruction technologies.

Chapter 4. Applications of data analytics for assessing PFAS exposure risks and source water impacts

4.1 Introduction

Improved methods to quantify PFAS in drinking water at lower levels has led to substantial changes in federal health advisories (Speth et al., 2022; Thagard, 2022). As mentioned previously, since 2016, the USEPA interim lifetime drinking water health advisory (LHA) for PFOA and PFOS combined had been 70 ng L^{-1} for the sum of the two concentrations (U.S. EPA, 2016b). In June 2022, interim LHAs were established for PFOA and PFOS individually and updated to 0.004 ng L^{-1} and 0.02 ng L^{-1}, respectively (U.S. EPA, 2022f). In addition to this change, the USEPA issued final health advisories for hexafluoropropylene oxide-dimer acid (HFPO-DA) and its ammonium salt (trade name, "GenX chemicals"), and perfluorobutanesulfonic acid and its related compound potassium perfluorobutane sulfonate (together referred to as "PFBS"): 10 ng L^{-1} for GenX and 2,000 ng L^{-1} for PFBS (U.S. EPA, 2022f).

Due to the complexity of PFAS issues, along with the dynamic nature of risk assessments and regulations, it is a challenge not only to keep up with information as it is generated (Crimi et al., 2022), but also, to clearly understand that information and its implications. This chapter demonstrates the utility of the approach developed in Chapter 3 to evaluate PFAS exposure risks as health guidance information continues to evolve. The UCMR3 PFAS data are used to spatially analyze exposure risks with respect to varying health benchmarks, investigate occurrence at facility and sampling point-level resolutions, and assess the impact of varying data aggregation methods on analysis outcomes. This work refines the current understanding of population exposure estimates, and identifies weaknesses in the UCMR data structure that will ultimately limit the accuracy of past and future population-wide exposure estimates. Methods to further utilization of the UCMR data by incorporating available supplemental data are explored and used to better the understanding of past temporal analyses of UCMR3 PFAS data.

4.2 Methods

4.2.1. Spatial analysis of exposure risks

For each PWS, average concentrations of PFAS above UCMR3 MRLs in samples from all EPs were calculated and used to compare these values to 1) the federal health guidance used by the USEPA to assess PFAS risk from May 2016 to June 2022 (70 ng L^{-1} in drinking water for the sum of PFOA and PFOS), and 2) the USEPA's most recent health advisories for PFOA and PFOS (0.004 ng L^{-1} for PFOA and 0.02 ng L^{-1} for PFOS). PFOA and PFOS concentrations were not reported in the UCMR3 dataset unless they were at least 20 ng L^{-1} and 40 ng L^{-1}, respectively. Nonetheless, the spatial analyses and visualizations presented here using both the former 70 ng L^{-1} LHA and the new interim LHAs demonstrate how the methods presented in this study can be applied to any risk benchmark. These methods will be especially beneficial in analyzing new monitoring data that have used improved analytical methods to quantify PFAS, such as recent statewide monitoring data and the pending UCMR5 data.

4.2.2. Evaluation of population-wide exposure estimates

To quantify populations potentially exposed to PFAS at the levels indicated by the UCMR3 data, the PWSID data field was used to link PWSs where samples exceeded benchmarks to populations of customers served using data available through the public

Safe Drinking Water Act (SDWA) Dataset (U.S. EPA, n.d.-a) using the XLOOKUP function in Microsoft Excel. The SDWA data has been used in previous assessments of population-wide exposure (Andrews & Naidenko, 2020; Hu et al., 2016).

The UCMR3 data structure was examined with respect to drinking water distribution to refine exposure estimates. As described in Chapter 3, the UCMR3 required quarterly sampling at each entry point (EP) to the distribution system for facilities using surface water (SW) and/or groundwater under the direct influence of surface water, and semi-annual sampling at EPs sourced by groundwater facilities (U.S. EPA, 2012b). The primary data fields used for these analyses are provided in Table 4.1.

Table 4.1. Primary data fields used.

Field Name	**Definition**
PWSID	9-character Public Water System (PWS) identification code
FacilityID	5-digit PWS facility identification code
FacilityWaterType	2-digit code identifying facility source water type: SW (surface water), GW (groundwater), GU (groundwater under the direct influence of surface water), MX (Any combination of SW, GW and GU)
SamplePointID	Identification code for each sample point location in the PWS
CollectionDate	Date of sample collection (month, day, year)
SampleID	Identification code for each sample, as defined by the laboratory
Contaminant	PFBS, PFHxS, PFHpA, PFOA, PFOS, or PFNA
AnalyticalResultsSign	“<” (less than MRL) or “=” (at or above the MRL)
AnalyticalResultValue	Analytical result in μg/L (null values represent concentrations < MRL)
Region	EPA region
State	State abbreviation

Figure 4.1 provides a process flow schematic for one of the PWSs monitored during UCMR3, a system serving residents of Florence, Alabama. As this example demonstrates,

a PWS may have one or more facilities which may be hydraulically independent treatment systems with unique raw water sources.

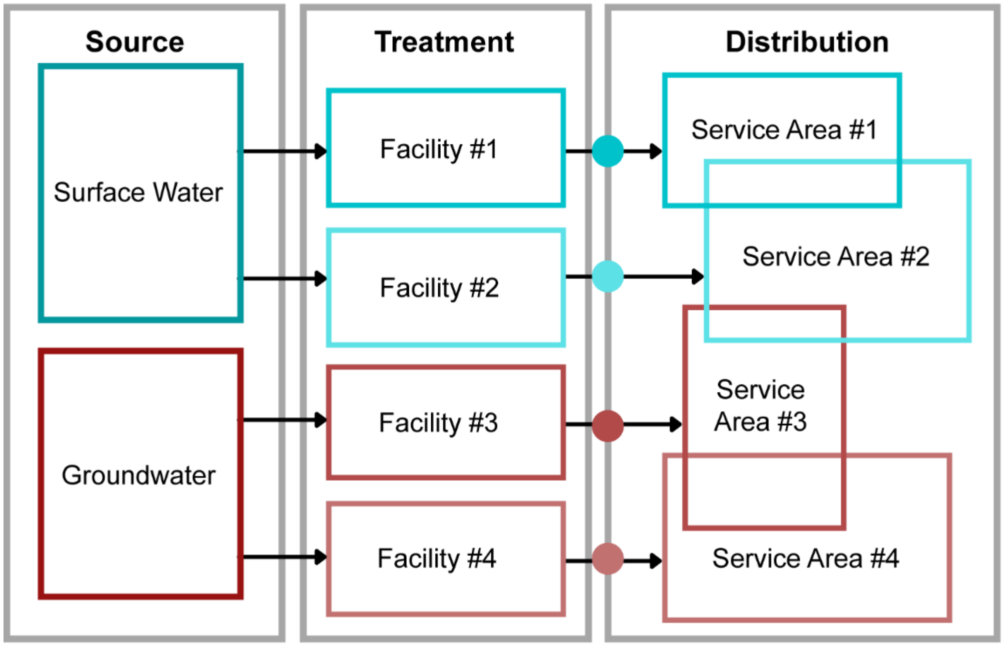


Figure 4.1. Public water system (PWS) process flow schematic.

Conceptual schematic of a northwest Alabama PWS monitored during UCMR3. Colored points indicate sampling points: discrete entry points (EPs) to the distribution network. Geographic areas of facility service areas are not provided in the UCMR3 occurrence database.

The PWS shown in Figure 4.1 collected a total of twelve samples throughout the UCMR3 program: four per surface water facility and two per groundwater facility. Figure 4.2 uses the PWS presented in Figure 4.1 to provide example data entries for attributes included with each PFAS record in the UCMR3 dataset. These data fields provide means to appropriately aggregate PFAS results by PWS, facility, and EP. With twelve samples collected (four per SW facility and two per GW facility) and analyzed for six PFAS, this PWS accounts for 72 records (0.03%) of the UCMR3 PFAS data. As discussed in Chapter 3 and shown in Figure 4.1, service areas associated with EPs are not provided in the UCMR occurrence data. The impact of this aspect of the occurrence data on exposure estimates was investigated by analyzing the data at facility and EP-levels, cross-referencing publicly available Consumer Confidence Reports (CCRs), and contacting utilities for further information.

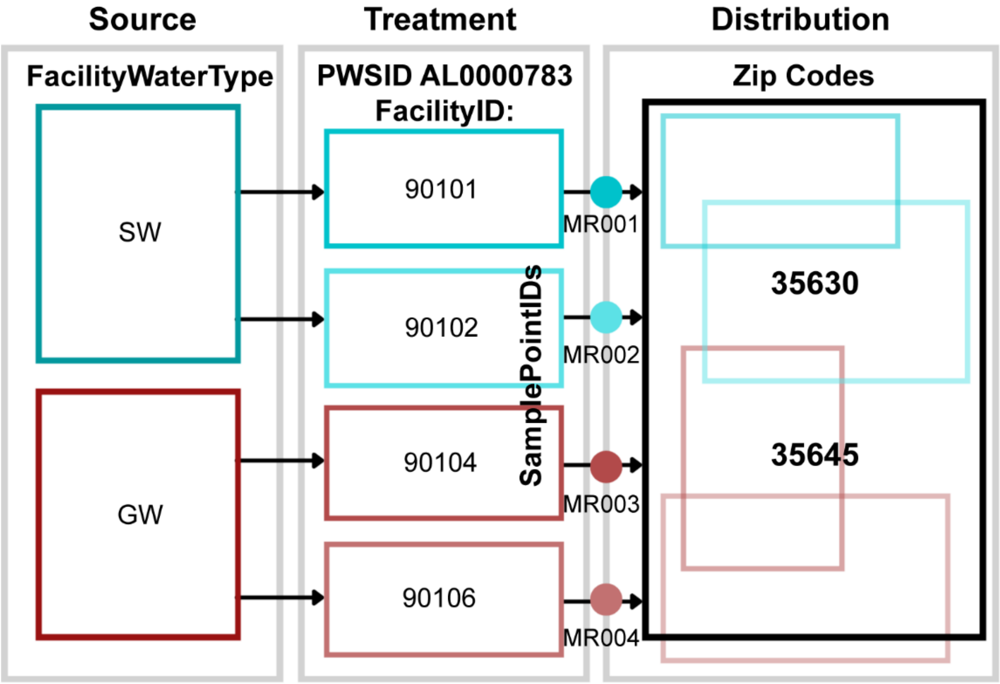


Figure 4.2. Example of UCMR3 data fields and corresponding values.

Schematic of a northwest Alabama PWS, labeled with UCMR3 data attributes and data field entries used as identifiers in data processing and aggregation. Zip code information associated with the dataset are also shown, illustrating the constraint on spatial resolution inherent in the dataset.

The development and execution of each UCMR round requires immense effort and investments from multiple parties including the individual drinking water utilities collecting samples (U.S. EPA, 2012b, 2021e). The data are both expansive and intricate, and with thousands of PWSs contributing to the overall database, slight variations in nomenclature from system to system must be accounted for in analyses. For example, the identification codes used by the PWS featured in the above figures clearly designate the system’s four facilities. The data associated with this PWS show that samples were collected and analyzed from each unique EP per the requirements of UCMR3. In the UCMR3 dataset at large, the “SamplePointID” is intended to designate the unique EPs sampled (U.S. EPA, 2017); however, validating the data obtained from this particular PWS revealed that the “SamplePointID” data field entries did not provide unique identifiers for each EP; thus, the “AssociatedSamplePointID” field served the purpose of the “SamplePointID,” and was reflected in this way in Figure 4.2. Had the data not been validated, a broad analysis of PFAS occurrence aggregated at the EP level would have produced one representative value for all four EPs as if all twelve samples were collected from the same location.

4.2.3. Investigation of further exploitation of UCMR data

For PWSs where PFAS are present and no advanced drinking water treatment technologies have been implemented to mitigate occurrence, there is evidence to suggest that differences between drinking water concentrations and environmental source water intake concentrations are not statistically significant (Boone et al., 2019; Glassmeyer et al., 2017). Therefore, the resolution of the nationwide, spatial occurrence data generated in this study allows for identification of regions and individual PWSs of interest for further investigation of PFAS behavior, fate, and impacts in source water. For example, the spatial analyses presented in this research reveal several PWSs with samples containing concentrations of PFAS above UCMR3 MRLs in North Carolina, Alabama, and Northwest Georgia. These sites were selected to investigate the applicability of UCMR3 data for furthering the understanding of PFAS impacts in source water by integrating data from supplemental sources, including both primary data (reconnaissance sampling data), and secondary data (information from the scientific literature and publicly available data from individual PWSs).

4.2.4. Temporal analyses

The data acquisition regime of the UCMR requires each PWS monitored during the three-year sampling effort to sample and report PFAS data for a one-year time frame. To investigate the effects of the spatiotemporal distribution of UCMR data density on assessments of temporal occurrence trends, the total number of PWSs monitored, samples collected, and PFAS concentrations detected were analyzed and spatially visualized by calendar year. Publicly available was collected from a PWS of interest, and the Mann-Kendall test was used to compare temporal trends at this location with the findings of a previous study that used the overall UCMR3 dataset to investigate the temporal occurrence trends (Guelfo & Adamson, 2018).

4.3 Results and Discussion

4.3.1. Exposure risks

The occurrence of PFOS and PFOA is visualized in Figure 4.3 as percent differences from the 2016 USEPA LHA in the continental United States. Following the methods presented in Chapter 3, PWS locations are based on the zip code centroids of the service areas for a given PWS. Overall, the analytical results of 197 samples exceeded the 2016 USEPA LHA of 70 ng L^{-1} for PFOA and PFOS combined. These samples represent 65 PWSs, 22 states, the Northern Marianas Islands, Guam, and the Salt River Pima-Maricopa Indian Community. Combined average concentrations of PFOA and PFOS were more than double the previous USEPA LHA (in effect from 2016-2022) at 27 PWSs within the continental U.S., one in Guam, and one in the Northern Marianas Islands.

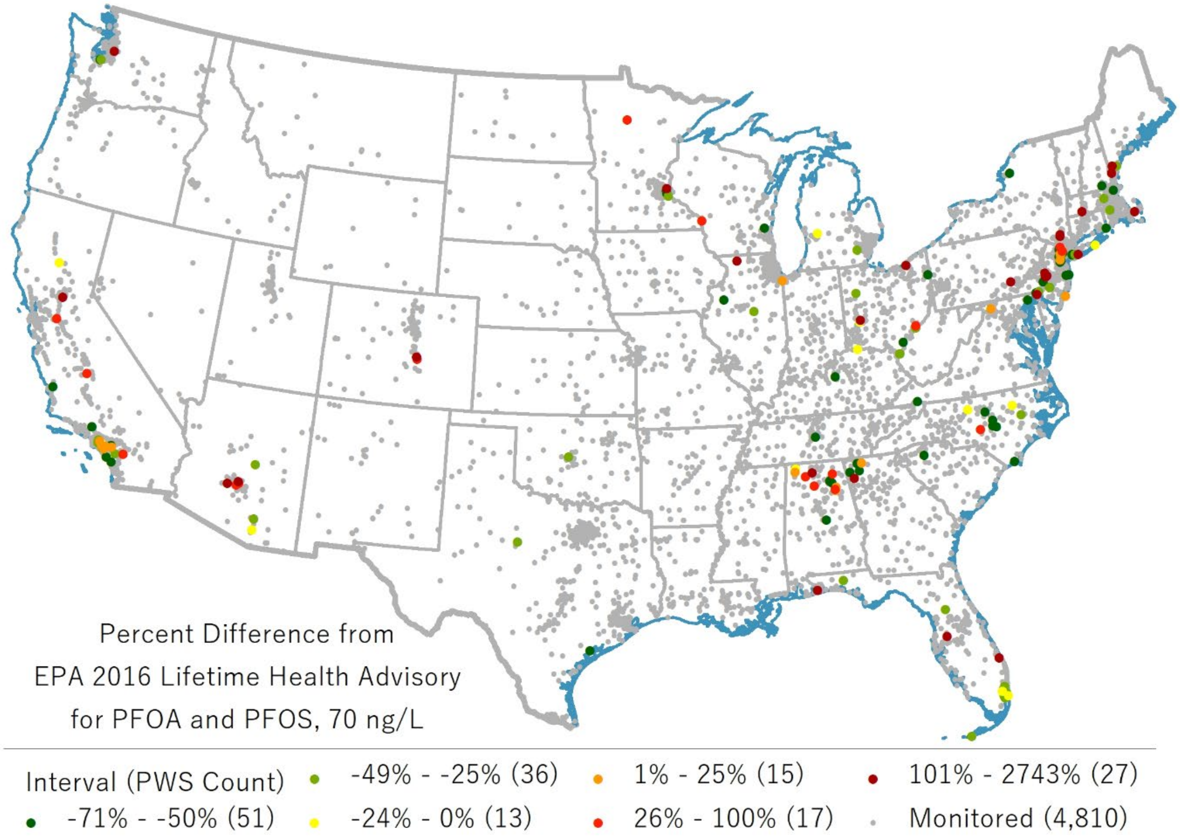


Figure 4.3. Nationwide PFOA and PFOS exposure with respect to 2016 USEPA lifetime health advisory, 70 ng L^{-1} for the sum of PFOA and PFOS

Points represent PWSs sampled during UCMR3. Colors indicate percent difference from the 2016 USEPA LHA of 70 ng L^{-1}, calculated using the total average concentrations of PFOA and PFOS exceeding MRLs at each PWS.

Although the results constitute a small percentage of total samples analyzed (0.5%) and PWSs monitored (1.3%), previous work by Hu et al. 2016 noted that approximately six million consumers are potentially provided drinking water by these 65 PWSs (Hu et al., 2016). Relative to the 2022 USEPA LHAs (0.004 and 0.02 ng L^{-1} for PFOA and PFOS, respectively), 508 (1.4%) samples analyzed during UCMR3 contained concentrations exceeding the current LHA. These samples represent 162 (3.3%) PWSs in 30 states, the

American Samoa, Guam, the Northern Marianas Islands, and Salt River Pima-Maricopa Indian Community.

The results of the PWS-level spatial analyses utilizing the 2022 USEPA interim LHAs for PFOA and PFOS are presented in Figure 4.4 and Figure 4.5, respectively. PWS locations are based on the zip code centroids of the service areas for a given PWS. Given the aforementioned difference between the reporting levels and the new interim LHAs for PFOA and PFOS, every PWS where at least one of these two compounds was reported at any concentration represents a substantial exceedance of current USEPA health guidance. States where PFAS concentrations were reported that, prior to June 2022, were understood to be below levels of human health concern, but that now must address extreme exceedances of new health advisories include Michigan, Kentucky, Texas, Oklahoma, South Carolina, and Tennessee.

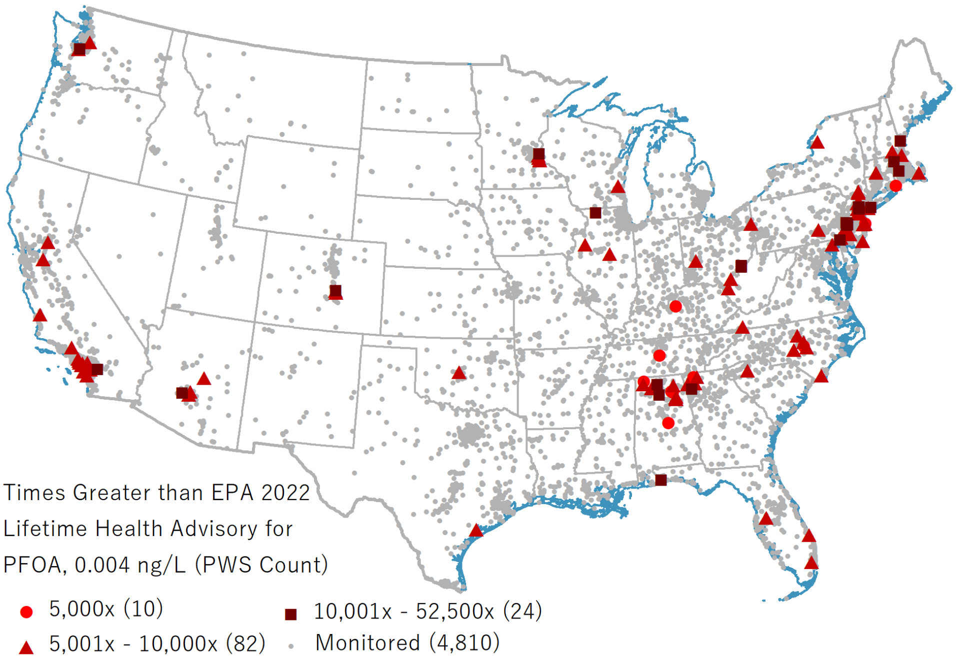


Figure 4.4. Nationwide PFOA exposure with respect to 2022 USEPA interim lifetime health advisory, 0.004 ng L^{-1}.

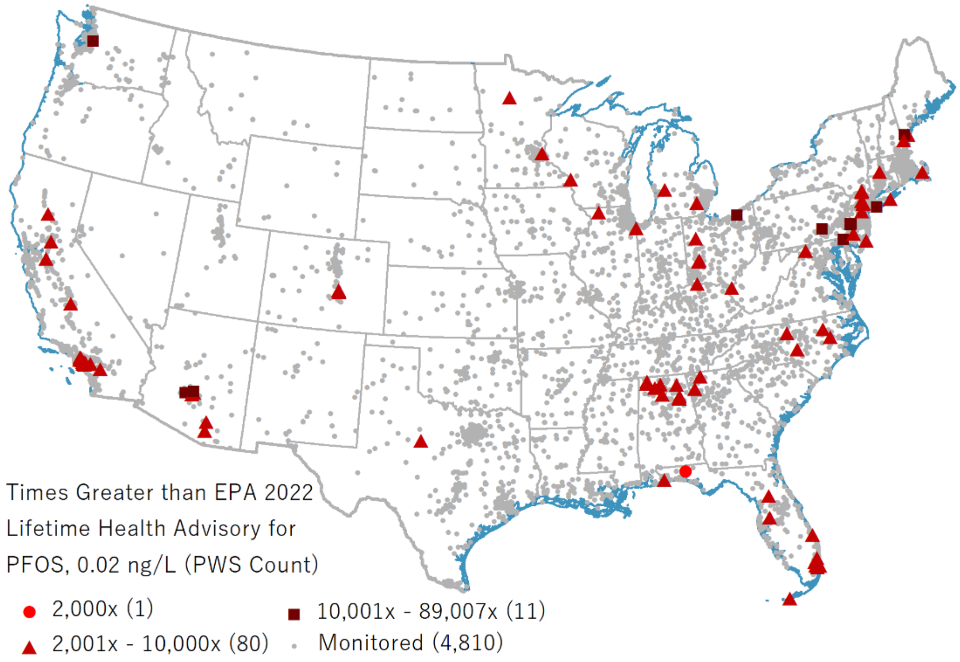


Figure 4.5. Nationwide PFOS exposure with respect to 2022 USEPA interim lifetime health advisory, 0.02 ng L^{-1}.

Using SDWA population data, the PWSs shown in Figure 4.4 and Figure 4.5 provide drinking water to approximately 14.8 million customers; however, there are several limitations embedded in this estimate which have implications for answering the question, "Do previous and future data management structures provide adequate information to accurately identify populations exposed to PFAS, and at what concentrations they are exposed?" The following section details key knowledge gaps related to population exposure: (1) distribution system service areas and operations, (2) duration of exposure, and (3) analytical quantitation limitations.

4.3.2. Population exposure uncertainty

This study analyzed the UCMR3 data at the finest spatial resolution possible, the PWS level, with PWS service area zip code centroids serving as a proxy for PWS location. The dataset contains spatial identifiers, i.e., the "SamplePointIDs" (the distribution system

EPs where samples were collected; Figures 4.1 and 4.2), that cannot be associated with exact geographic service areas given the current available data. Moreover, because EPs are related to the actual distribution networks that move finished water to end-users, it is not possible to accurately determine the actual number of potential consumers exposed to PFAS-contaminated drinking water. With these constraints in mind, coupling EP-level insights with PWS-level spatial analyses provides an approach to more fully understand the UCMR3 results and limitations; to refine the current understanding of the extent of PFAS exposure and estimates of populations affected; and to examine the effects of data aggregation on analysis outcomes. In the following sections, the utility of integrating EP-level and PWS-level analyses into future UCMR PFAS studies is demonstrated by examining UCMR3 results in southeastern New York. The average *reported* concentrations at each PFAS are provided in Figure 4.6. Refer to Figure 3.2 regarding aggregation methods and visualization.

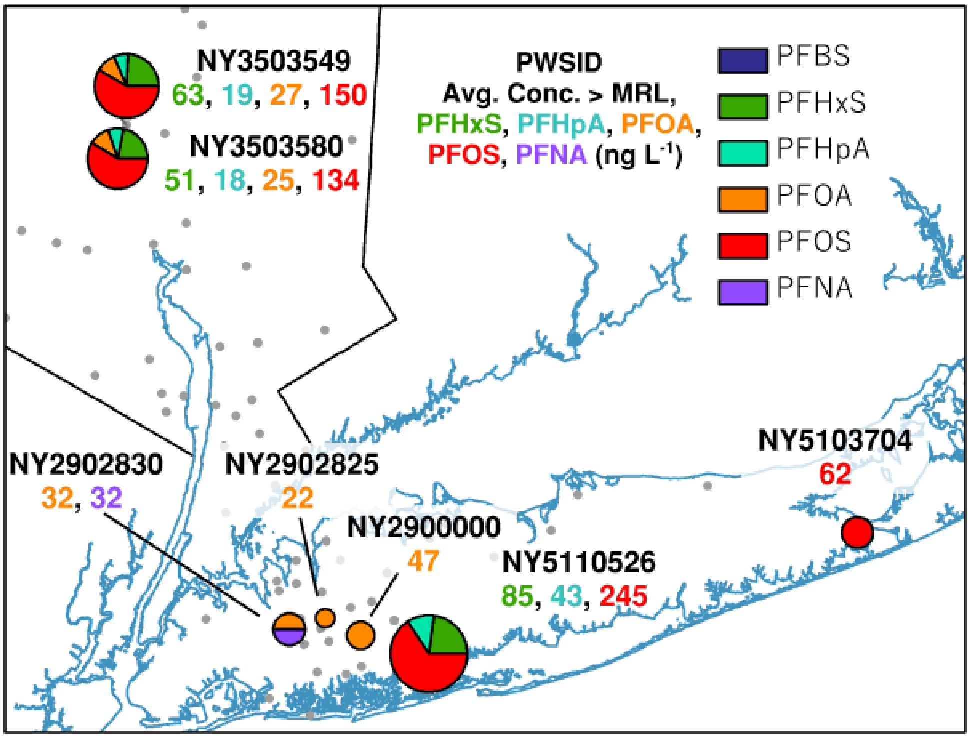


Figure 4.6. UCMR3 results exceeding minimum reporting levels in the Long Island, New York area.

Grey circles indicate PWSs that were monitored, but where no PFAS were reported at any EP.

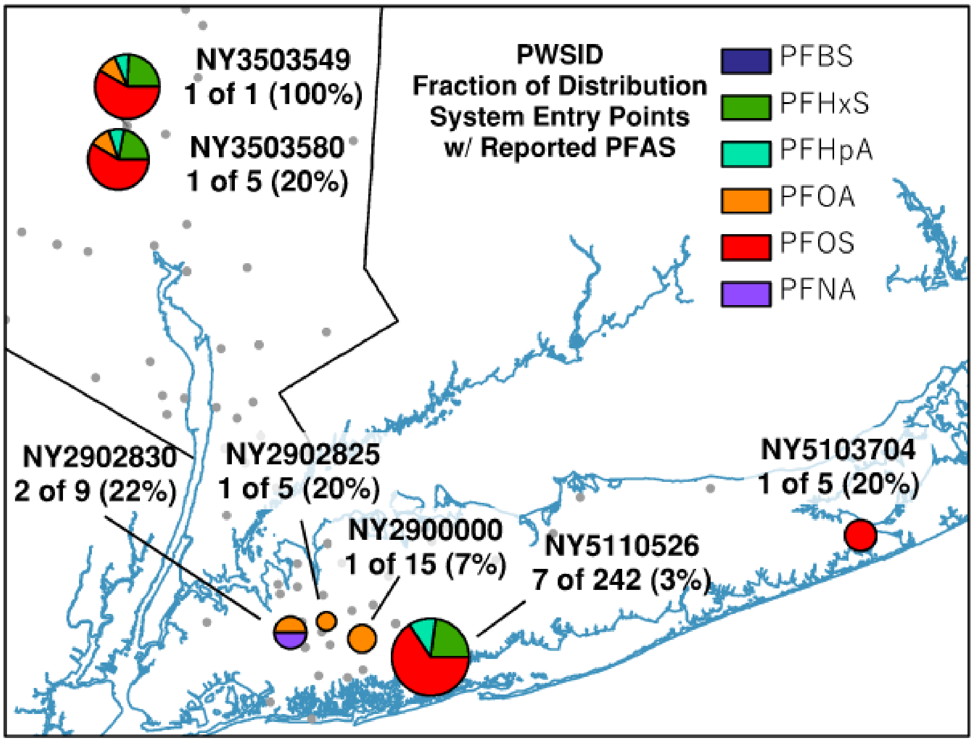


Figure 4.7. Fraction of distribution system entry points represented by UCMR3 results exceeding minimum reporting levels in the Long Island, New York area.

Grey circles indicate PWSs that were monitored, but where no PFAS were reported at any EP.

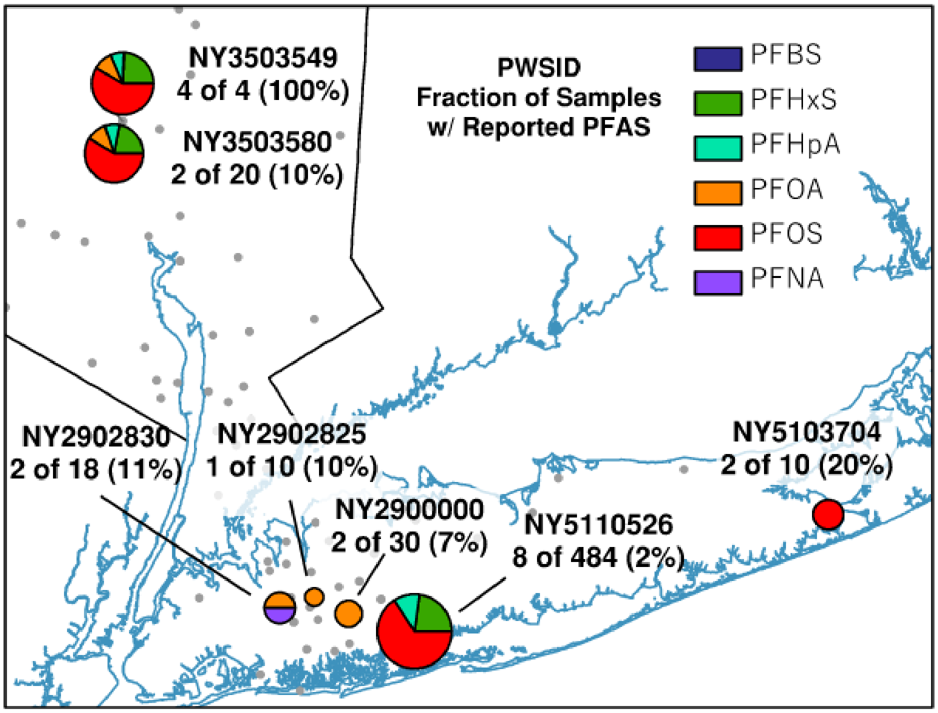


Figure 4.8. Fraction of samples represented by UCMR3 results exceeding minimum reporting levels in the Long Island, New York area.

Grey circles indicate PWSs that were monitored, but where no PFAS were reported at any EP.

Uncertainty due to distribution system entry points, service areas, and operations

Many PWSs reported PFAS contamination at a subset of their total EPs. Within the 198 PWSs where at least one PFAS was reported in at least one sample, 1,673 distribution system EPs were sampled. One or more PFAS were reported at only 267 (16%) of these EPs. The total number of distribution system EPs for the 4,920 individual PWSs sampled during UCMR3 ranged from 1 (e.g., Figure 4.7, NY3503549) to 242 (Figure 4.7, NY3503549). Of the 198 PWSs where PFAS contamination was reported, 149 (75%) are comprised of systems with more than one EP to the distribution network. The large majority of these PWSs with multiple EPs and PFAS concentrations exceeding MRLs (138, 93%) reported PFAS concentrations exceeding MRLs at only some of their EPs. For these

PWSs, using the total population served by the PWS to estimate populations at risk of PFAS exposure overestimates the true population exposed.

In other words, the UCMR3 dataset indicates that 49 (25%) of the participating PWSs with reported concentrations of PFAS collected and analyzed samples from only one EP (e.g., NY3503549) which indicates that the PWS either has only one EP or that the selected EP was approved by the USEPA as representative of all EPs to their distribution system (refer to Section 3.2.1). This implies the entire population served drinking water by each of these PWSs may be at risk of PFAS exposure. Of the subset of PWSs with reported concentrations of PFAS, an additional 11 PWSs sampled more than one EP and PFAS were reported at every EP. Similarly, the entire population served by these PWSs may be at risk of PFAS exposure at the levels indicated by the UCMR3. However, using the total population served by every PWS to estimate population-wide exposure for the remaining 138 PWSs with PFAS exceeding MRLs at only some of their EPs (e.g., all PWSs in Figure 4.7, except NY3503549) artificially inflates populations exposed at the levels indicated by the UCMR3.

For example, SDWA data indicate that PWS NY5110526 provides drinking water to approximately 1.1 million residents (U.S. EPA, n.d.-a) and contributed the largest quantity of data to the UCMR3 dataset: 484 samples from 242 EPs (1.3% of all UCMR3 data). Comparatively, equal representation across all 4,920 PWSs sampled would result in each PWS contributing 0.02% of the dataset. Unequal data distribution is discussed further in Section 4.3.5. At this PWS, 7 (2.9%) of the 242 EPs sampled, and 8 (1.7%) of the 484 samples analyzed had concentrations of PFAS exceeding MRLs. Previous studies have used these SDWA data, which represents total population served by an entire PWS, to estimate population-wide exposure (Andrews & Naidenko, 2020; Hu et al., 2016). However, these estimates are limited in that they do not consider that the vast majority of entry points to end-users did not report any concentrations of PFAS. Previous assessments of exposure and risk also neglect the distribution network design and operations for each unique PWS.

To refine the current understanding of populations at risk of exposure to PFAS, and to what extent, 22 representative PWSs were contacted to inquire about distribution system details. Representatives from 10 PWSs responded, and 5 indicated that either all or the majority of their facilities were capable of serving all of their customers. However, they were unable to provide details about the volume of water from various entry points as it is combined in the distribution system to serve their customers. The remaining PWSs provided varying degrees of information, or none at all, claiming that distribution system information is proprietary. Two PWSs reported facilities serving discrete zones of customers but did not provide estimates of customers served by each facility. One PWS reported two facilities with overlapping customer zones. Conversations with representatives from these PWSs revealed two factors that complicate efforts to improve estimates of populations exposed to PFAS through drinking water: the proprietary nature of some drinking water distribution networks and the dynamic nature of the networks (e.g., wells can be taken on or offline based on demand).

Uncertainty due to duration of exposure

The USEPA has indicated that lifetime health advisories (LHAs) "indicate levels to protect all people, including sensitive populations and life stages, from adverse health effects resulting from exposure *throughout their lives* to these PFAS in drinking water. The health advisory levels were calculated to offer a margin of protection against adverse health effects. EPA's lifetime health advisories also take into account other potential sources of exposure to these PFAS beyond drinking water (for example, food, air, consumer products, etc.), which provides an additional layer of protection" (U.S. EPA, 2022e), emphasis mine. Thus, it is important to consider whether UCMR3 results indicate populations being *consistently* exposed to PFAS, and at what levels. For example, Table 4.2 shows results of all sampling events at groundwater-sourced EPs where PFAS were reported in the state of New York. Notably, at NY5110526 (where 242 EPs were sampled), PFAS exceeded MRLs during only one of the two sampling events at 6 of the 7 EPs with reported concentrations of PFAS. At the 1 EP where PFAS exceeded MRLs in both samples, the concentrations differed substantially between the two sampling events (Table 4.2, Facility ID 643).

Table 4.2. UCMR3 results (ng L^{-1}) at entry points (EPs) sourced by groundwater facilities where at least one PFAS concentration was reported in New York.

PWS ID Facility ID PFAS	Reported PFAS Concentrations in Drinking Water at Entry Points Sourced by Groundwater Facilities in New York (ng L^{-1})											
	2013				2014				2015			
	1	2	3	4	5	6	7	8	9	10	11	12
NY2212214												
90001												
PFHpA							30		30			
PFHxS							70		80			
PFOA							30		40			
NY2900000												
8956												
PFOA						48			45			
NY2902825												
7512												
PFOA				▨		22						
NY2902830												
2414												
PFNA								32			▨	
7649												
PFOA								32			▨	
NY5103704												
95224												
PFOS							82			41		
NY5110526												
304												
PFHpA					▨			14				
PFHxS					▨			85				
PFOS					▨			150				
309												
PFHxS						▨	43					
338												
PFOS											▨	44
362												
PFHpA					▨		16					
PFHxS					▨		98					
PFOS					▨		330					
636												
PFHpA		▨		60								
643												
PFHxS			60					140				
PFOS			170					530				
649												
PFHpA	▨			82								

▨ <MRL

PFHxS

PFHpA

PFOA

PFOS

PFNA

Results at entry points (EPs) sourced by surface water facilities where at least one PFAS concentration was reported are provided in Appendix Table A.4.

The UCMR3 PFAS data in New York show that where multiple wells are present, PFAS occurrence across EPs and over time is not consistent in frequency or concentration (Table 4.2). Therefore, for PWSs comprised of multiple treatment facilities with service areas related to a variety of EPs, using the total population served by the entire PWS does not accurately reflect the true population exposed. For PWSs where service areas associated with various EPs overlap (see Figure 4.1), and only some EPs contain PFAS above reporting levels, the concentrations may be diluted in the distribution network; therefore, the risk of exposure at the concentrations represented may overestimate true exposure.

Analytical and reporting uncertainty

As discussed in Section 3.2.3, the high MRLs of the UCMR3 data limit present a major limitation to infer information about the low-level, chronic exposure that many populations may experience through drinking water sources. However, the methodology presented here may be applied to existing statewide monitoring efforts and the upcoming UCMR5 data, which utilize newer analytical methods and substantially lower reporting levels.

4.3.3. Assessing aggregation and analysis methods

Further investigation of PWS NY5110526 and the HUC-8 watershed it is located within (HUC 02030202, Southern Long Island) demonstrates dataset nuances that may lead to misinterpretation of broad occurrence statistics previously reported in the literature and highlights how varying aggregation methods and analyses affect interpretation of UCMR results. For example, in the study performed by Hu et al. (2016), a spatial regression model was developed to predict PFAS concentrations in HUC-8 watersheds based on the number of possible PFAS sources within them. The authors used a single sample, the sample with the maximum concentration, to represent each HUC-8 watershed (Hu et al., 2016). The total number of samples collected from PWSs within the southern Long Island watershed is 831. The aggregation method used to create the spatial regression model skews the PFAS concentrations used in the analysis upward because it does not account for any data associated with any of the other 830 samples collected within the watershed. Therefore, the visualizations presented by Hu et al. (2016) provide worst-case contamination scenarios at the watershed-level scale. The spatial methods presented in this study focus on drinking water occurrence and exposure risks at the PWS-level. In the case of the Southern Long Island subbasin, 23 samples were used to generate averaged data for eight PWSs in this analysis and visualization, rather than one maximum value being used to represent an entire subbasin. Figure 4.9 illustrates the difference between the aggregation methods of the work of Hu et al. (2016) and the present study.

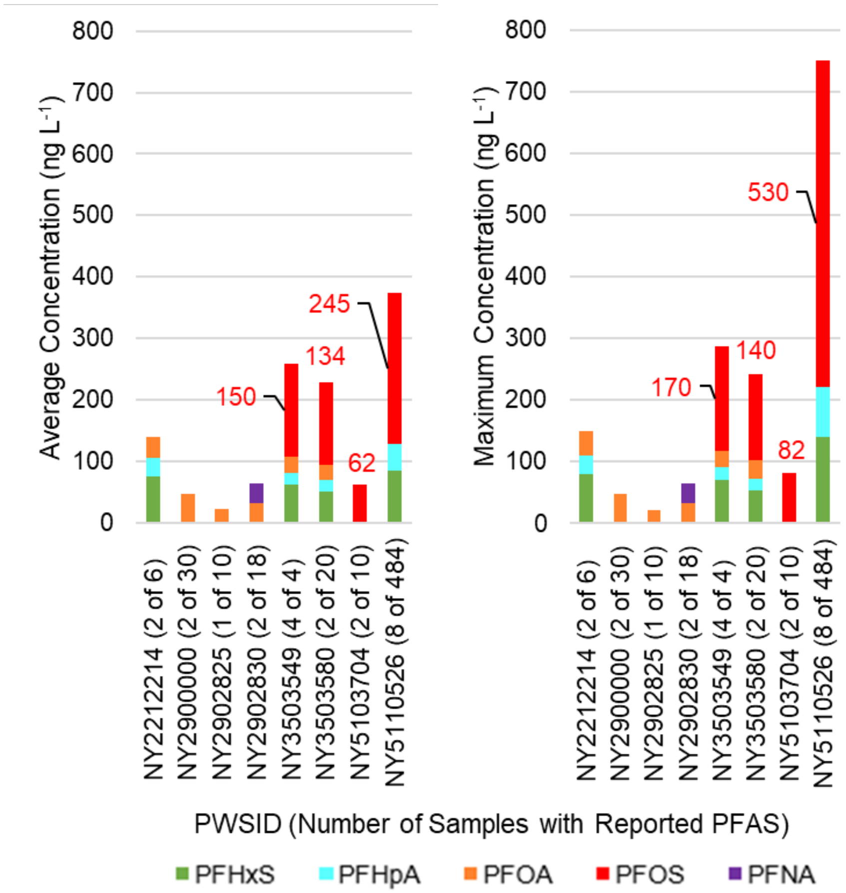


Figure 4.9. PWS-level UCMR3 results within HUC 02030202 (Southern Long Island Watershed)

Numbers in parenthesis following PWSIDs indicate number of samples exceeding USEPA-determined MRLs for at least one PFAS.

Figure 4.9 provides the average and maximum concentrations of PFAS reported above UCMR3 MRLs for each PWS within the HUC-8 watershed. The 530 ng L^{-1} maximum contribution of PFOS that occurred at NY5110526 is also the maximum concentration of PFOS detected in USEPA Region 2 throughout the duration of the UCMR3 (Table 3.6). This value was used by Hu et al. (2016) to represent the HUC-8 watershed. In contrast, the methods presented in this work spatially visualize each of the eight PWSs shown in Figure 4.9 using zip code centroid points, rather than aggregating the results for the eight PWSs at the HUC-8 watershed-level. The point representing PWS NY5110526 in all spatial analyses presented in this research (Chapter 3 and Chapter 4) represents the average of the five samples exceeding the PFOS MRL at the PWS, 245 ng L^{-1}.

4.3.4. Expanding utility of UCMR data

PFAS contamination in the Cape Fear River of North Carolina has previously been reported in the literature (Sun et al., 2016). Figure 4.10 provides an example of how integrating data from various sources can inform research directions to better understand the occurrence of PFAS in both drinking water and source water. In Figure 4.10a, the spatial distribution of PFAS occurrence in drinking water at discrete PWSs across the HUC-8 Cape Fear Basin is visualized to evaluate how PFAS constituents and concentrations in drinking water varied across the basin. Figure 4.10b provides a comparison between UCMR3 drinking water data (Figure 4.10a) and existing source water intake data by visualizing the approximate locations of three PWS surface water intakes where samples were collected and analyzed for PFAS during the timeframe of the UCMR3 (Sun et al., 2016). These surface water samples, collected and analyzed for seventeen PFAS analytes by Sun et al. (2016), revealed a suite of PFAS in surface water within the Cape Fear River watershed, with ten PFAS exceeding the study reporting levels at the PWS intake furthest upstream (mean ΣPFAS = 355 ng L^{-1}), nine at the intake of the PWS near the middle of the basin (mean ΣPFAS = 62 ng L^{-1}), and eight at the PWS intake furthest downstream (mean ΣPFAS = 710 ng L^{-1}) (Sun et al., 2016). The sum of the average concentrations of the seventeen PFAS analyzed by Sun et al. are labeled in Figure 4.10b. Only the six PFAS analyzed during UCMR3 are visualized in pie charts for comparison.

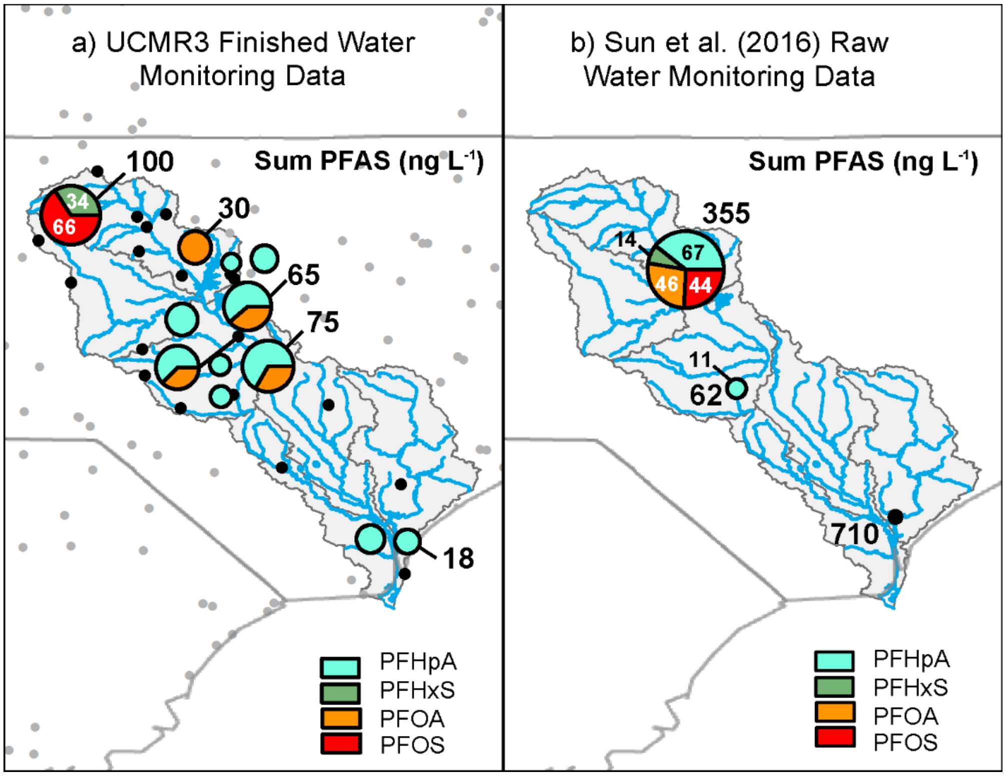


Figure 4.10. PFAS Occurrence in Cape Fear Watershed, North Carolina.

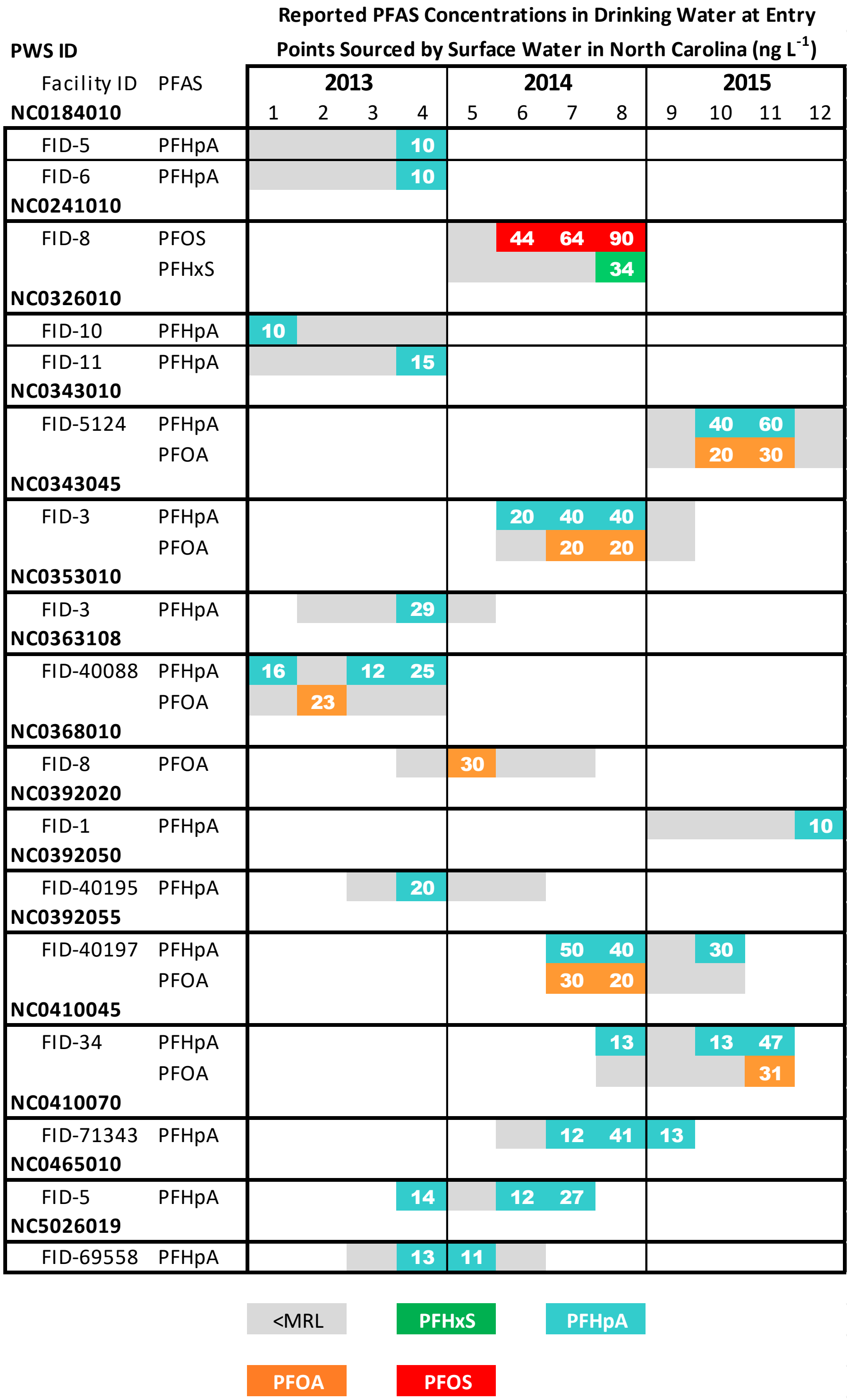
Concentrations of UCMR3 PFAS analytes in (A) finished drinking water, as provided by UCMR3 data, and (B) surface water, as provided by the work of Sun et al. (2016). Size of pie charts scale with sum of UCMR3 PFAS analyte concentrations. Contributing concentrations of individual UCMR3 analytes are provided for PWSs with maximum total PFAS concentrations.

(A) Average concentrations of PFAS in finished drinking water from PWSs sourced by surface water sampled during UCMR3.

(B) Average concentrations of PFAS in source water sampled from system intakes by Sun et al. (2016) during the UCMR3 timeframe. Sum PFAS quantities account for the total suite of PFAS analyzed in the 2016 study (Sun et al., 2016).

The sum of the mean individual PFAS concentrations for each source water site (Figure 4.10b) are substantially higher than those found in the drinking water samples during the UCMR3, but the samples collected and analyzed by Sun et al. (2016) targeted a total of seventeen PFAS analytes: ten legacy PFAS and seven novel perfluoroalkyl ether carboxylic acids (PFECAs), as opposed to the six analytes monitored during UCMR3 (Sun et al., 2016). While UCMR3 drinking water sampling was more robust in geographic reach, the sampling efforts of Sun et al. (2016) at the three source water locations were much more robust in number of samples collected and number of PFAS analyzed. Sun et al. (2016) collected 127 samples at the headwaters of the watershed, and median concentrations of PFHxS, PFHpA, PFOA, and PFOS were 10 ng L^{-1}, 39 ng L^{-1}, 34 ng L^{-1}, and 29 ng L^{-1}, respectively. Although median results for these analytes were similar to those reported across the watershed during UCMR3, at this intake, concentrations ranged as high as 193 ng L^{-1} (PFHxS), 324 ng L^{-1} (PFHpA), 137 ng L^{-1} (PFOA), and 346 ng L^{-1} (PFOS). Sun et al. (2016) analyzed 73 samples at the intake in the middle of the watershed, and the median concentrations of the six PFAS that were analyzed during UCMR3 (PFBS, PFHxS, PFHpA, PFOA, PFOS, and PFNA) were below the quantitation limits of the study (<10 ng L-1 for PFBS, PFHxS, PFHpA, PFOA, and PFNA; <25 ng L^{-1} for PFOS). This finding suggests that the lack of PFBS and PFNA occurrence, and the infrequent occurrence of PFOS and PFHxS (Table 4.3), may not be due to the higher MRLs used for the UCMR3 data. At this intake, the maximum concentration of PFHpA was 85 ng L^{-1} and the most frequently detected PFAS were two that were not analyzed in UCMR 3, perfluorobutanoic acid (PFBA) and perfluoropentanoic acid (PFPeA). At the source water intake furthest downstream, the maximum concentration of PFHpA was 24 ng L^{-1}, which is comparable to the average concentrations reported nearby during UCMR3. At this location, however, the concentration of perfluoro-2-propoxypropanoic acid (PFPrOPrA), the ammonium salt of which is a known PFOA alternative, ranged from 55 to 4,560 ng L^{-1}.

Table 4.3. UCMR3 results (ng L^{-1}) at entry points (EPs) sourced by surface water facilities where at least one PFAS concentration was reported in North Carolina.



Reported PFAS Concentrations in Drinking Water at Entry Points Sourced by Surface Water in North Carolina (ng L^{-1})

PWS ID Facility ID	PFAS	2013				2014				2015			
		1	2	3	4	5	6	7	8	9	10	11	12
NC0184010													
FID-5	PFHpA	▨	▨	▨	10								
FID-6	PFHpA	▨	▨	▨	10								
NC0241010													
FID-8	PFOS					▨	44	64	90				
	PFHxS					▨	▨	▨	34				
NC0326010													
FID-10	PFHpA	10	▨	▨	▨								
FID-11	PFHpA	▨	▨	▨	15								
NC0343010													
FID-5124	PFHpA									▨	40	60	▨
	PFOA									▨	20	30	▨
NC0343045													
FID-3	PFHpA						20	40	40	▨			
	PFOA						▨	20	20	▨			
NC0353010													
FID-3	PFHpA		▨	▨	29	▨							
NC0363108													
FID-40088	PFHpA	16	▨	12	25								
	PFOA	▨	23	▨	▨								
NC0368010													
FID-8	PFOA				▨	30	▨	▨					
NC0392020													
FID-1	PFHpA									▨	▨	▨	10
NC0392050													
FID-40195	PFHpA			▨	20	▨	▨						
NC0392055													
FID-40197	PFHpA							50	40	▨	30		
	PFOA							30	20	▨	▨		
NC0410045													
FID-34	PFHpA								13	▨	13	47	
	PFOA								▨	▨	▨	31	
NC0410070													
FID-71343	PFHpA						▨	12	41	13			
NC0465010													
FID-5	PFHpA				14	▨	12	27					
NC5026019													
FID-69558	PFHpA			▨	13	11	▨						

▨ <MRL   PFHxS   PFHpA

PFOA   PFOS

Results at entry points (EPs) sourced by groundwater facilities where at least one PFAS concentration was reported are provided in Appendix Table A.5.

In Alabama, 124 PWSs participated in UCMR3, representing 416 facilities of all source water types: surface water (SW), groundwater (GW), groundwater under the influence of surface water (GU), and mixed sources (MX). Most facilities were categorized as either SW (106, 25%) or GW (303, 73%) facilities. There were 6 and 1 GU and MX facilities, respectively. Of the 416 facilities represented by the data, PFAS were reported at 24 facilities (6%). Despite the large proportion of GW-sourced facilities sampled, the majority of drinking water samples with reported concentrations of PFAS were collected from SW-sourced facilities (20, 83%). Figure 4.11 shows the number of PFAS records exceeding MRLs at each PWS, and Figure 4.12 displays the average reported concentrations of PFAS in North Alabama and Northwest Georgia.

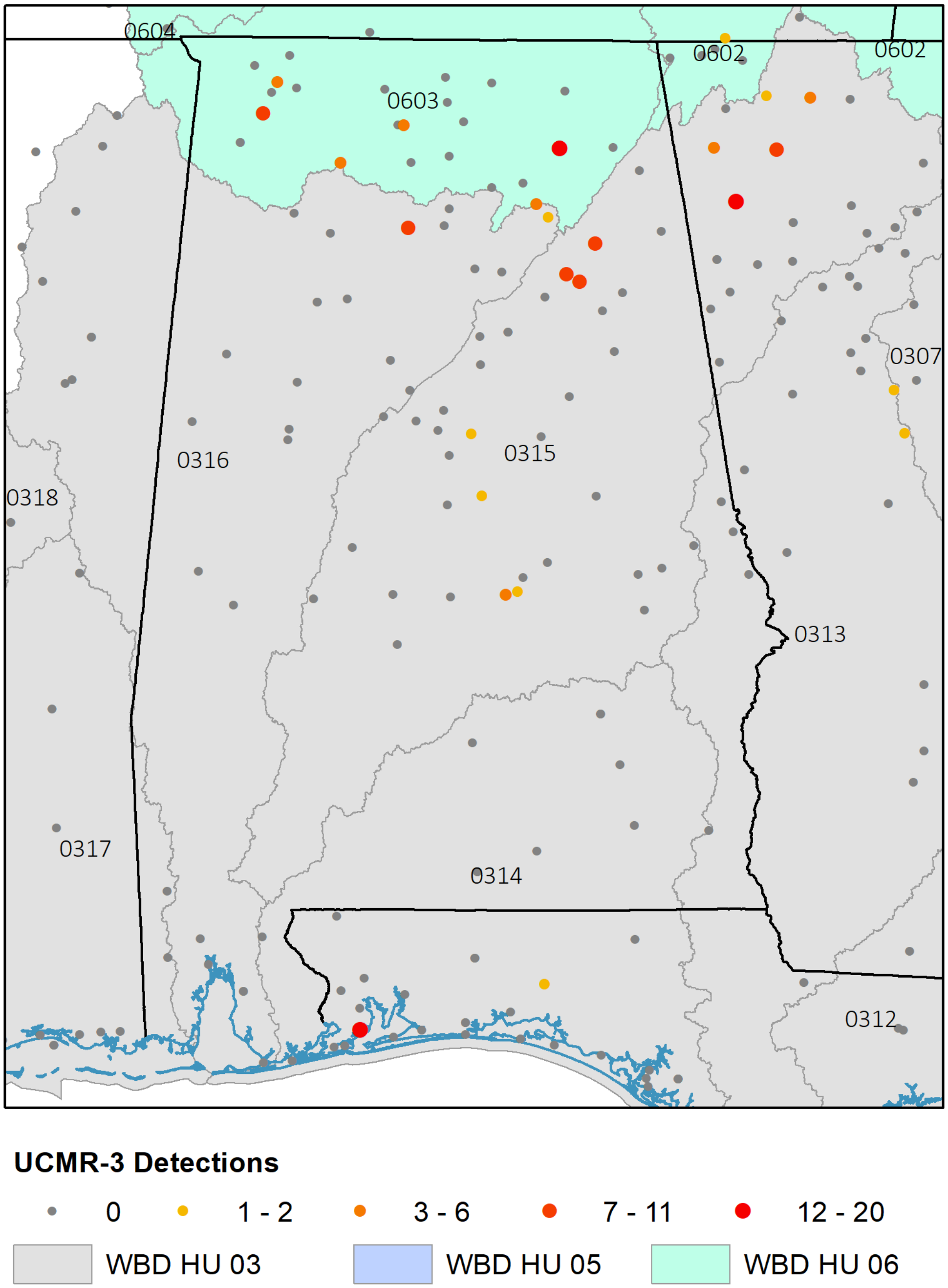


Figure 4.11. Number of PFAS records exceeding MRLs in Alabama 4-digit hydrologic unit codes (HUC-4).

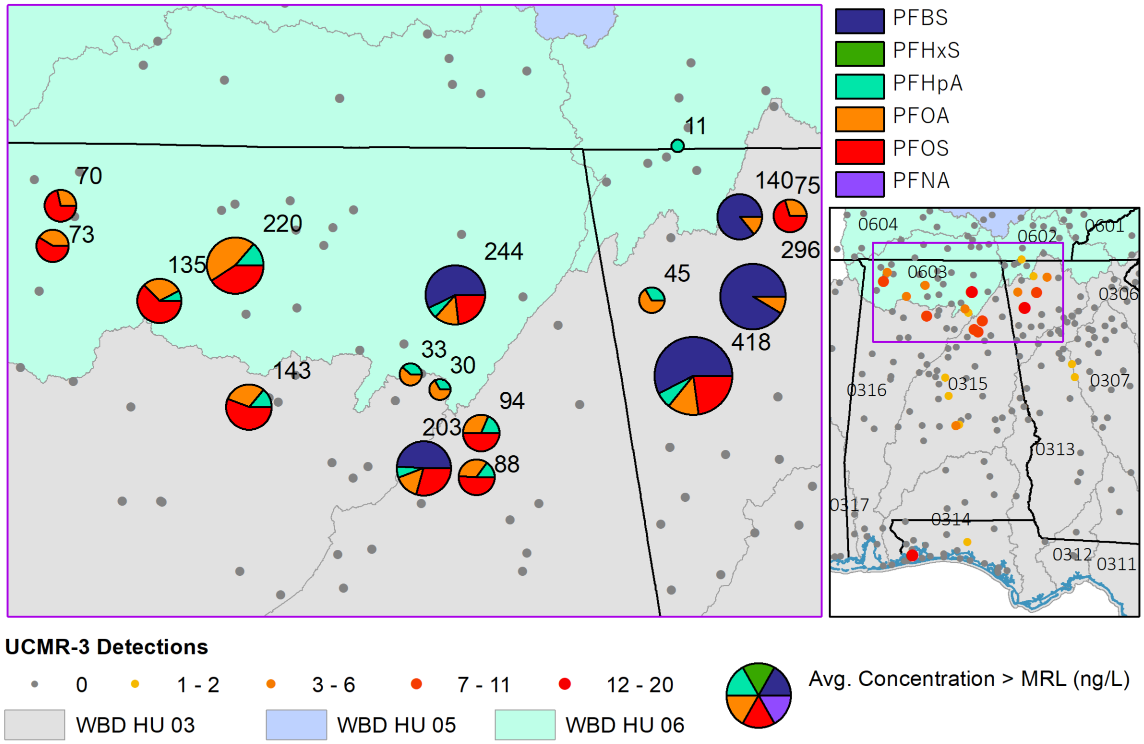


Figure 4.12. Average reported concentrations of PFAS in North Alabama and Northwest Georgia.

In a 2020 statewide survey of the spatial distribution and mass transport of seventeen PFAS in Alabama surface water (including all UCMR3 analytes), the limit of detection did not exceed 1.8 ng L^{-1} for any analyte, yet only six PFAS were detected: PFBS, PFPeA, PFHxA, PFHpA, PFOA, and PFOS (Viticoski et al., 2022). Concentration trends in the Coosa and Alabama River basins follow closely with overall PFAS occurrence during UCMR3 (Figure 4.11, HUC 0315). Interestingly, within the Tennessee River basin (Figure 4.11, HUC 0603), the maximum $\sum_6$ PFAS concentration detected in any sample was 35.6 ng L^{-1}, and PFHpA was not detected in any sample. Comparing the UCMR3 samples collected from 2013-2015 (Figure 4.11) with the findings of Viticoski et al. (2022) could suggest that the 2002 phase-out of PFOS and gradual phase-out of PFOA starting in 2006 have reduced occurrence of these two compounds within the Tennessee River;

however, it is also quite possible that the technologies implemented to manage PFAS in this region as a result of UCMR3 sampling, and the litigation with local fluorochemical manufacturers that followed, have decreased the overall PFAS load in this watershed (3M, 2022; Thornton, 2019).

The UCMR3 data can also be used by researchers to investigate PFAS occurrence in areas where no PFAS were reported. In a collaborative reconnaissance sampling effort performed in 2018, samples were collected from Tennessee and North Carolina and analyzed for ten PFAS using the LC-MS/MS methods described by Viticoski et al. (2022). Despite being collected from different watersheds and separated by a broad geographic distance, results were similar across all samples with respect to both ∑PFAS and constituent contribution (Figure 4.13).

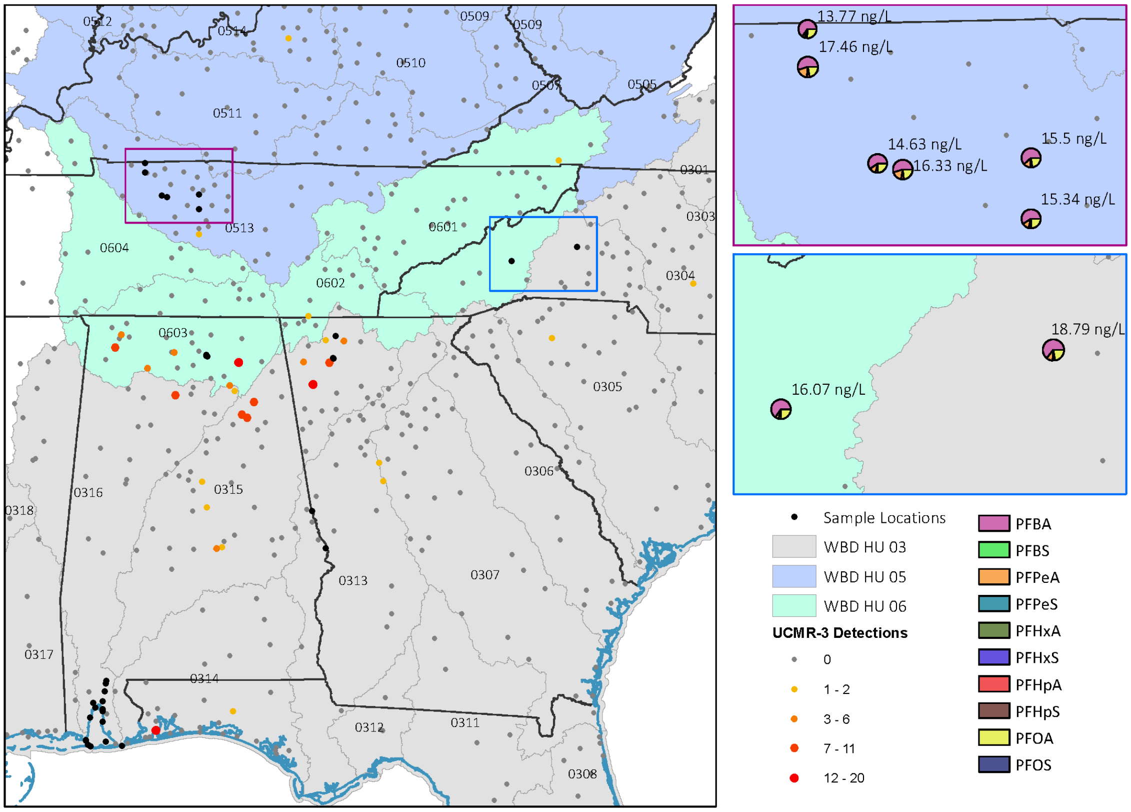


Figure 4.13. Reconnaissance sampling results in Tennessee and North Carolina.

The lower reporting levels and larger number of PFAS analyzed in these studies can be leveraged to better understand PFAS occurrence in source water and to identify PFAS that are likely present in drinking water. As low concentrations of PFAS become increasingly relevant to human health and the list of total PFAS of concern continues to grow, coupling data available from both environmental and drinking water studies provides an avenue for informing research directions and remediation priorities. UCMR data serve as useful indicators for PFAS in the environment and may enlighten strategic planning for future environmental monitoring efforts, but there is no central database relating PWSs to their specific environmental source water. However, consulting publicly available Consumer Confidence Reports (CCRs) of systems of interest provides an avenue for gaining information regarding PWS and facility-level source water (Appendix Figures A.1 and A.2). For example, consulting CCRs in areas of interest revealed that PWSs in Northwest Georgia reporting PFAS occurrence during UCMR3 were sourced by the Conasauga, Etowah, and Oostanaula Rivers, among others (Appendix Figure A.2). Primary and secondary data acquisition allowed for historic trends in PFAS occurrence and constituent distribution in source water to be investigated. In 2006, long-chain PFAS (PFOA, PFOS, and PFNA) were substantial contributors to the overall PFAS load (Appendix Figure A.3), but short-chain PFAS were not analyzed (Konwick et al., 2008). In samples collected two years later (Appendix Figure A.4), overall concentrations of PFAS had decreased, and a suite of short-chain PFAS had become the predominant PFAS contaminants (Lasier et al., 2011). A decade later (in 2019), primary reconnaissance sampling efforts revealed similar overall PFAS loads, but a single short-chain constituent, PFBS, was the primary contributor (Appendix Figure A.6), which was consistent with drinking water occurrence findings through UCMR3 in samples collected in 2015 (Figure 4.12).

4.3.5. Temporal analyses

A 2018 study used the 36,578 UCMR3 samples collected during the official rule duration (2013-2015) to calculate quarterly occurrence frequencies and investigate the hypothesis that temporal trends in PFAS occurrence would reflect their recalcitrance, as well as changes in use and source control (Guelfo & Adamson, 2018). This study provided moderate evidence of increasing trends in overall PFAS occurrence frequency, but did not provide evidence that the 2002 phase-out of PFOS and gradual phase-out of PFOA starting in 2006 had an impact on occurrence frequencies during the UCMR3 monitoring period (Guelfo & Adamson, 2018). Spatial analysis at the PWS-level provides context for better understanding these results, and for understanding the limitations of broad analyses of temporal trends in PFAS occurrence without also accounting for spatial variation in the UCMR3 data.

Because the UCMR3 requirements were based on PWS size and facility source water type and only required each PWS to participate for one year, the resulting dataset does not provide equal representation in data quantity across space and time. Figure 4.14, Figure 4.15, and Figure 4.16 provide spatial visualizations of PWSs sampled each calendar year of the official UCMR3 timeframe (2013-2015) and the number of samples collected at each PWS for that year. Each sample distribution map is paired with a map visualizing the spatial distribution of overall PFAS occurrence for that year, displayed as the sum of the average reported concentrations of each analyte (ng L^{-1}).

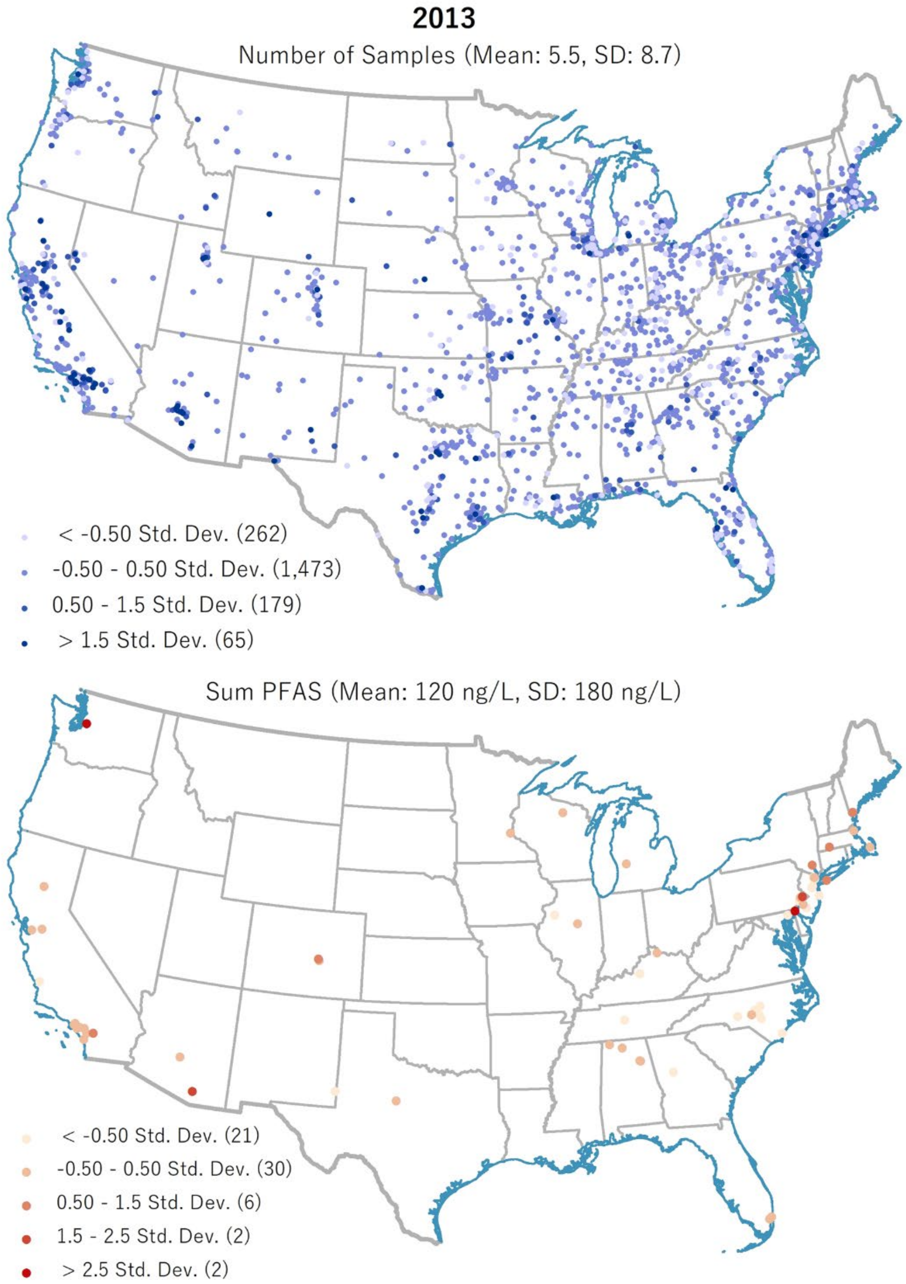


Figure 4.14. Spatial distribution of samples collected in 2013 and total concentrations of PFAS reported.

Sum PFAS represents the total of the average concentrations of each of the six PFAS analytes reported over UCMR3 MRLs in 2013.

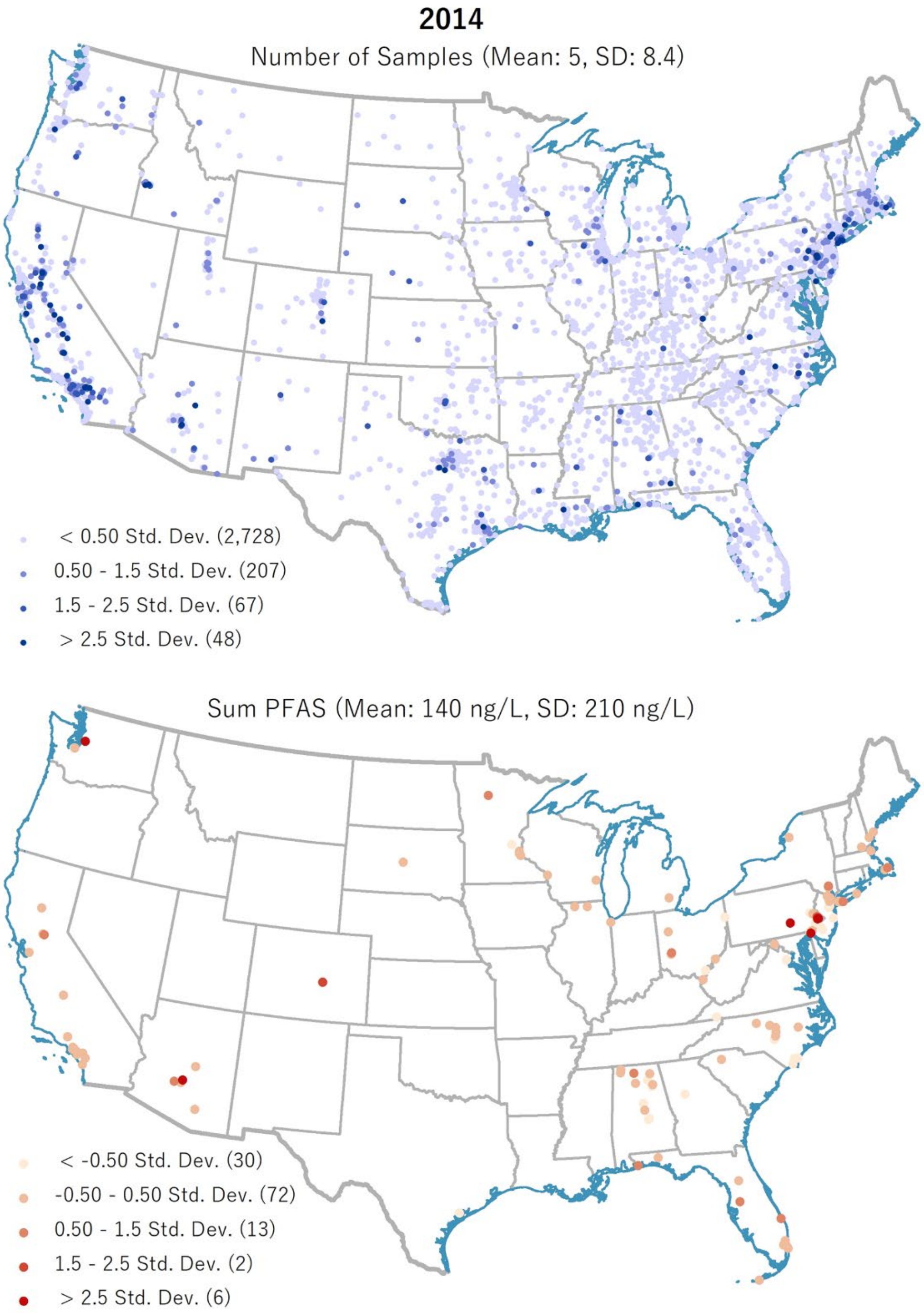


Figure 4.15. Spatial distribution of samples collected in 2014 and total concentrations of PFAS reported.

Sum PFAS represents the total of the average concentrations of each of the six PFAS analytes reported over UCMR3 MRLs in 2014.

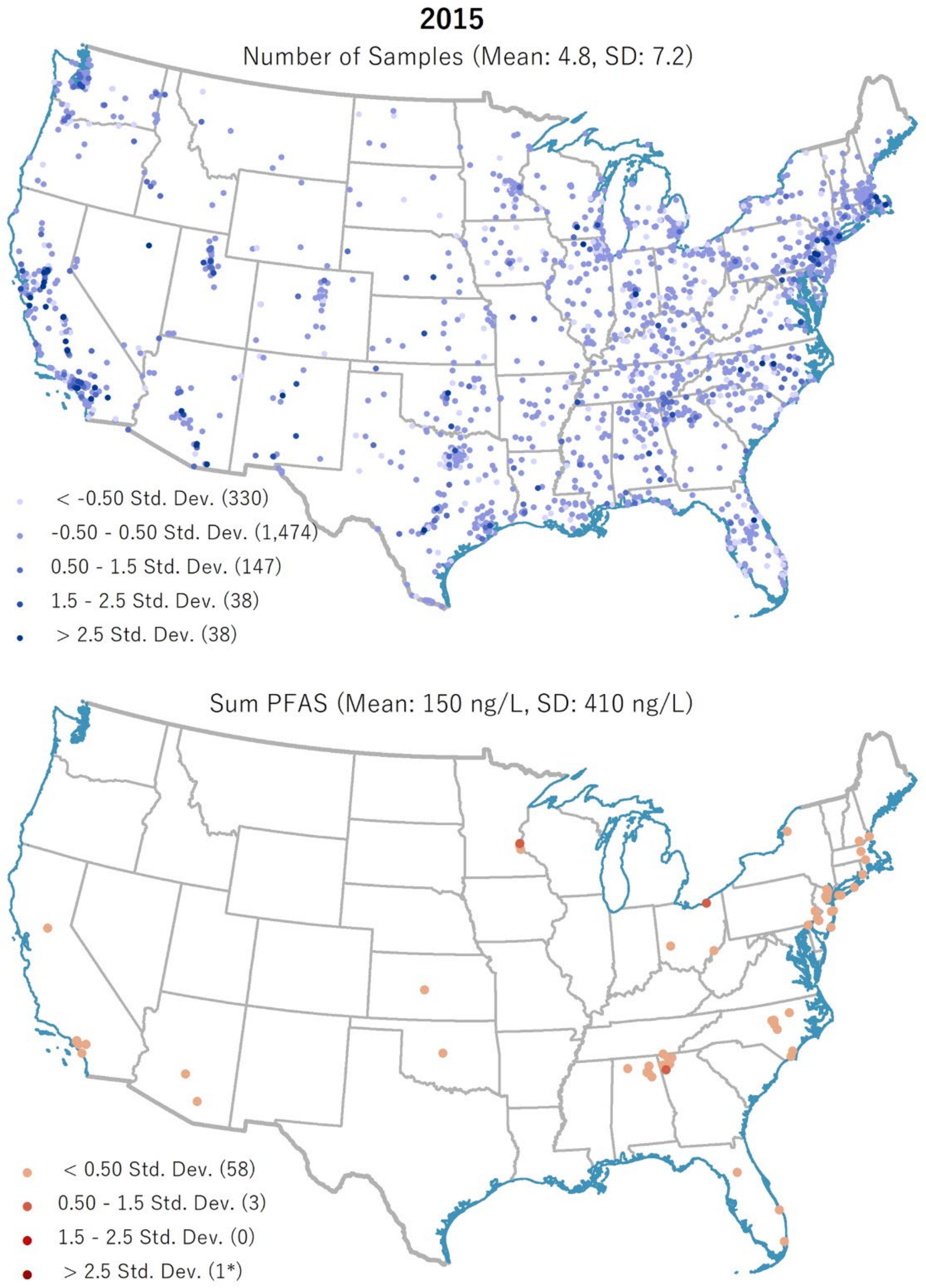


Figure 4.16. Spatial distribution of samples collected in 2015 and total concentrations of PFAS reported.

Sum PFAS represents the total of the average concentrations of each of the six PFAS analytes reported over UCMR3 MRLs in 2015.

The above figures display the variation in UCMR3 data density distribution over the official three-year duration of UCMR3, which introduces uncertainty in broad analyses of temporal occurrence trends. For example, 50% of all UCMR3 samples can be attributed to nine states, with California and Texas alone accounting for 25% of the data (17% and 8%, respectively). Temporal analyses using the nationwide data collectively obscures regional trends where PFAS occurrence is more prevalent. For example, in California (which represents 17% of all UCMR3 data), PFAS were reported during 11 of the 12 quarters of the UCMR3 sampling duration (no PFAS exceeded MRLs during the twelfth quarter, during which less than 2% of California's total samples were collected). In contrast, for the entire UCMR3 duration, PFAS were reported in only three samples in Texas (which accounts for 8% of all UCMR3 data), and no PFAS were reported after the fifth sampling quarter. Broadly temporally analyzing samples from areas with few PFAS sources and very low occurrence frequencies together with samples from areas where PFAS occurrence frequency is greater could mask trends in occurrence at PFAS hotspots that could aid in understanding the effects of PFAS recalcitrance, use, and source control on drinking water occurrence. To explore this hypothesis, publicly available data for PFOA and PFOS occurrence in source water from a PWS within the Coosa River basin (AL0000577) was collected from the Alabama Department of Environmental Management (ADEM) eFiles (ADEM, 2022a).

Following the same methodology of Guelfo et al. (2018), the Mann-Kendall test was used as a non-parametric assessment of temporal trends in PFAS concentration over time using ProUCL 5.2.0 (U.S. EPA, 2022d). Analyzing the publicly available source water data from April 2016 to February 2022 (Figure 4.17) revealed statistically significant evidence of a decreasing trend in PFOA concentrations ($p<0.001$) and insufficient evidence of a significant trend in PFOS concentrations (Appendix Figures A.7 and A.8). These findings differ from the results of the Mann-Kendall tests performed by Guelfo et al. of nationwide occurrence frequencies (aggregated quarterly), which indicated an increasing trend in PFOA occurrence ($p=0.01$), and a probable increasing trend in PFOS occurrence ($p=0.1$).

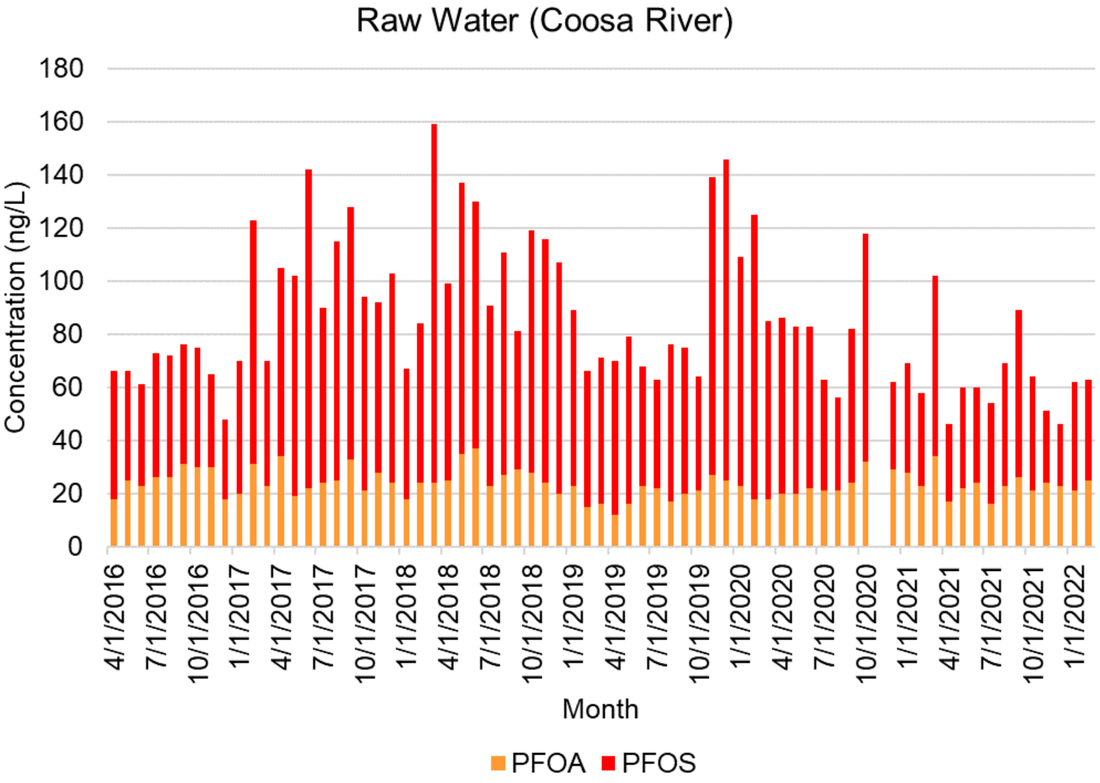


Figure 4.17. Routine monitoring data from Alabama PWS with reported concentrations of PFAS during UCMR3.

4.4 Conclusions

This work details the nuances of UCMR data collection, quantitative analyses, and data structure; furthers the current understanding of nationwide PFAS exposure trends; and provides methods for obtaining new insights specific to stakeholder needs. This study demonstrates the utility of past and future UCMR data for identifying geographic hotspots where significant levels of PFAS have been detected and for quantifying worst-case-scenario nationwide exposure. Additionally, the EP-level aggregation and analysis methods presented here can be applied to increase the understanding of PFAS occurrence phenomena within individual public water systems. To the best of the author's knowledge, this study is the first to provide exposure assessments for PFOA and PFOS using the USEPA LHAs issued in June 2022. This work emphasizes nuances of UCMR monitoring and database management; presents a straightforward method for visualizing PFAS exposure risks that can be used as the scientific understanding of human health risks improves and regulatory actions increase; and provides an approach for stakeholders to fill knowledge gaps pertinent to their specific PFAS monitoring and remediation needs. This approach will be applicable as new data become available and as new health and regulatory benchmarks are determined. As Mehan (2021) described on behalf of the AWWA, there are existing tools that the USEPA could be using to a greater degree to help address PFAS concerns in the water industry (Mehan, 2021). This work provides an accessible methodology for scientists and stakeholders to more fully utilize a USEPA-provided tool, the UCMR data, to advance the current understanding of PFAS occurrence and exposure risks.

Chapter 5. Conclusions

5.1 Impact

Given the complex and evolving nature of PFAS challenges, and the resources required to research and manage PFAS occurrence in source and drinking water systems, utilization of existing data are vital to accelerating the current scientific understanding of PFAS at a rate that allows stakeholders to effectively and expediently address risks associated with exposure. These approaches foster the exploitation of existing data to guide investigative and remediation efforts, design relevant studies and experiments, and inform regulatory decisions. In 1970, Tobler argued that “everything is related to everything else, but near things are more related than distant things” (Tobler, 1970). This concept, which has proliferated in the field of geography and is at the core of spatial analysis and modeling (Miller, 2004), has profound implications for progressing the state of the science of PFAS. The spatial component of PFAS data is essential for deriving meaning from analytical results and taking effective action. Spatial analyses are paramount in identifying vulnerable populations, planning future monitoring endeavors, focusing remediation efforts, and enlightening regulatory decisions. Therefore, advances in analytical technology to detect and quantify PFAS in drinking water and the environment must be matched with systematic data management and analysis methodologies to understand the implications of the results.

This work illustrates the utility of the UCMR for understanding the human health and ecological implications of PFAS occurrence in the U.S., and the potential and power of straightforward data analysis and GIS approaches to feasibly guide future PFAS research. Many factors cause difficulty in keeping pace with PFAS-related knowledge demand. In addition to an ever-growing list of PFAS (Wang et al., 2017), analytical challenges (Guelfo et al., 2018; Kotthoff & Bücking, 2018), and unknown environmental behavior, fate, and transformation pathways (Ahrens & Bundschuh, 2014); social, political, and economic influences further complicate our scientific understanding of PFAS (Brennan et al., 2021; Cordner et al., 2019). The comprehensive data acquisition efforts needed to address these challenges are costly and not always feasible. Exploiting existing and pending data to its fullest extent allows for accelerated assessments of the impacts of PFAS occurrence in source and drinking water, a clearer understanding of exposure risks, strategic investigative and remediation efforts, relevant experimental design, and informed action plans.

5.2 Limitations, recommendations, and future directions

Establishing best practices in database management and analysis is essential as greater quantities of PFAS data are produced. In future assessments of UCMR PFAS occurrence data and exposure risks, the geographic distribution of data density across PWSs, the wide range of EPs represented by individual PWSs, varying facility source water types, and overall PWS operations should be accounted for. Given the nature of the program requirements, the holistic dataset is more representative of PWSs with large distribution networks, i.e., many EPs, rather than PWSs with smaller networks and fewer EPs. Furthermore, the dataset assigns greater weight to PWSs with facilities sourced by surface water, since four samples are mandated per EP at these facilities, compared with the two required at groundwater facilities. Therefore, broad assessments of nationwide occurrence frequencies and population-wide exposure, without refined analyses, yields potentially misleading results to readers unfamiliar with UCMR monitoring efforts and occurrence data.

For accurate interpretation of existing and future PFAS monitoring datasets, data limitations must be clearly understood. In the case of the UCMR, the lack of information regarding the relationship between sample collection points (EPs) and distribution to end users limits population exposure estimates (in both number of customers exposed, and the levels at which they are exposed). Joining SDWA population data with UCMR data is useful in quantifying populations *potentially* exposed to PFAS at the levels indicated, but without information about the operation of PWSs, including service areas and approximate number of customers served by discrete EPs, more realistic population-wide exposure estimates are not possible. These considerations will be important for interpreting the upcoming UCMR5 data, since the final rule does not indicate that EP-level spatial or customer information will be provided (U.S. EPA, 2021e). The relatively high reporting levels used in the UCMR3 program also limited assessments of occurrence trends across time and space. The recent USEPA health advisories and final rule for the UCMR5 indicate that MRLs will again be a limitation in assessing the extent of exposure at levels of human health concern (U.S. EPA, 2021e, 2022f). Lastly, the UCMR3 was limited by the total number of PFAS analyzed, as six of a growing family of thousands of PFAS were monitored. Because the UCMR5 will analyze these six and an additional twenty-three PFAS at lower reporting levels, these data will help to fill critical knowledge gaps related to the current understanding of PFAS occurrence.

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Appendix

Table A.1. UCMR3 data field names and definitions (U.S. EPA, 2017)

Field Name	Definition
PWSID	Public Water System Identification Code, 9-character identification code (Begins with the standard 2-character postal State abbreviation or Region code, and the remaining seven numbers are unique to each PWS in the state)
PWSName	Name of the Public Water System (PWS)
Size	Size category of the PWS for UCMR, based on retail population as of December 31, 2010: S (≤ 10,000), L (> 10,000)
FacilityID	Public Water System Facility Identification Code, 5-digit identification code
FacilityName	Name of the facility at the PWS
FacilityWaterType	Source of water at the facility: SW (surface water), GW (ground water), GU (ground water under the direct influence of surface water), MX (Any combination of: SW, GW and GU)
SamplePointID	Identification code for each sample point location in the PWS
SamplePointName	Name of the sample point for every sample point ID at a PWS
SamplePointType	Sampling Point Type Code: EP (entry point to the distribution system), MR (distribution system at maximum residence time)
AssociatedFacilityID	The facility ID of the associated MR
AssociatedSamplePointID	The sample point ID of the associated MR

Field Name	**Definition**
Disinfectant Type	CLGA (Gaseous Chlorine), CLOF (Offsite Generated Hypochlorite, stored as liquid), CLON (Onsite Generated Hypochlorite, no storage), CAGC (Chloramine, formed from gaseous chlorine), CAOF (Chloramine, formed from offsite hypochlorite), CAON (Chloramine, formed from onsite hypochlorite), CLDO (Chlorine Dioxide), OZON (Ozone), ULVL (Ultraviolet Light), OTHD (All other types of disinfectant), NODU (No Disinfectant Used)
CollectionDate	Date of sample collection (month, day, year)
SampleID	Identification code for each sample, as defined by the laboratory
Contaminant	Unregulated contaminant being analyzed in UCMR 3
MRL	Minimum Reporting Level defined by UCMR 3 in µg/L for the chemicals
MethodID	Identification code of the analytical method
AnalyticalResultsSign	Less than (<) the minimum reporting level (MRL) or equal to (=) a numeric value at or above the MRL
AnalyticalResultValue	Numeric value of the analytical result in µg/L for the chemicals, null values represent less than MRL
SampleEventCode	Identification code for each sample event. Includes sample event one (SE1), sample event two (SE2), sample event three (SE3), and sample event four (SE4).
MonitoringRequirement	AM (Assessment Monitoring, List 1), SS (Screening Survey, List 2), PST (Pre-Screen Testing, List 3)
Region	EPA Region (States): 1 (CT, ME, MA, NH, RI, VT), 2 (NJ, NY, PR (Puerto Rico), VI (Virgin Islands)), 3 (DE, DC, MD, PA, VA, WV), 4 (AL, FL, GA, KY, MS, NC, SC, TN), 5 (IL, IN, MI, MN, OH, WI), 6 (AR, LA, NM, OK, TX), 7 (IA, KS, MO, NE), 8 (CO, MT, ND, SD, UT, WY), 9 (AZ, CA, HI, NV, AS (American Samoa), GU (Guam), MP (Northern Marianas Islands), NN (Navajo Nation)), 10 (AK, ID, OR, WA)
State	State abbreviation

Field Name	Definition
ZipCode	U.S. Postal Service zip code(s) for all areas being served water by a PWS

Table A.2. Manually rectified zip code validation errors.

PWSID	Zip Code Listed	Rectified Zip Code Centroid Used for Spatial Analysis	
		Latitude	Longitude
90605013	92953	33.507379	-117.12176
AS9711948	96799	-14.27342	-170.70303
AZ0407033	85242	33.2528	-111.5239
AZ0411017	85232	33.067703	-111.47051
AZ0411060	85242	33.24815	-111.63449
AZ0411086	85272	32.879341	-111.96315
FL6410322	33561	-	-
GU0000010	96540	13.444304	144.79373
IL0310780	60635	41.923672	-87.809807
IL1450250	61888	38.011494	-89.23925
MP0000001	96950	15.194839	145.7473
NC0416015	28857	34.729301	-76.750587
OH3102612	45228	39.066384	-84.427477
PA5260036	15602	40.216621	-79.601921
SD4600141	57357	43.137134	-98.426378
TN0000273	37747	36.171997	-82.825493
WA5300119	V4W2X3	49.060604	-122.49276

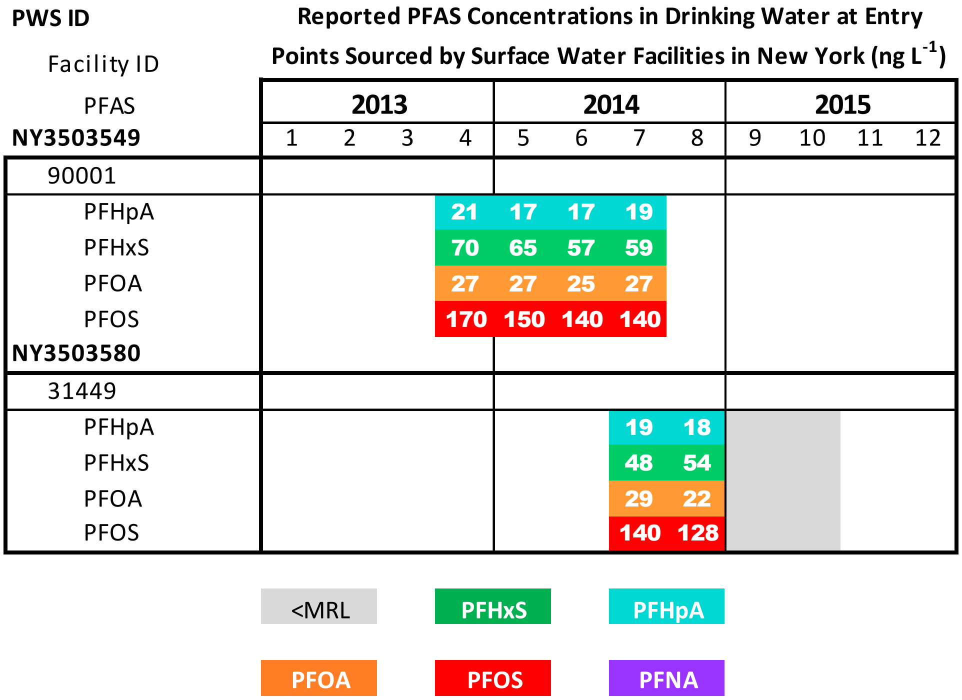
Table A.3. Total number of analytical results, number of results exceeding MRLs, and frequency of detections above MRLs for each region and state/territory/associate.

EPA Region	**Number of Analytical Results**	**Number of Analytical Results > MRL**	**% > MRL**
State/Territory			
1	**11598**	**63**	**0.54**
1	48	0	0
CT	2100	0	0
MA	7248	47	0.65
ME	492	5	1.02
NH	750	8	1.07
RI	672	3	0.45
VT	288	0	0
2	**22212**	**174**	**0.78**
NJ	8766	122	1.39
NY	10104	51	0.5
PR	3282	1	0.03
VI	60	0	0
3	**14226**	**150**	**1.05**
DC	96	0	0
DE	780	38	4.87
MD	1362	1	0.07
PA	8160	91	1.12
VA	3012	2	0.07
WV	816	18	2.21
4	**41621**	**239**	**0.57**
AL	6336	89	1.4
FL	8760	50	0.57
GA	4529	39	0.86
KY	3702	4	0.11
MS	3684	0	0
NC	7920	53	0.67
SC	2778	2	0.07
TN	3912	2	0.05
5	**25266**	**66**	**0.26**
5	72	0	0
IL	7728	5	0.06
IN	2622	2	0.08

EPA Region State/Territory	**Number of Analytical Results**	**Number of Analytical Results > MRL**	**% > MRL**
MI	4296	3	0.07
MN	2664	31	1.16
OH	3978	17	0.43
WI	3906	8	0.2
6	**27936**	**12**	**0.04**
6	36	0	0
AR	1620	0	0
LA	3432	0	0
NM	2022	1	0.05
OK	3078	4	0.13
TX	17748	7	0.04
7	**7410**	**1**	**0.01**
IA	1470	0	0
KS	1296	1	0.08
MO	3384	0	0
NE	1260	0	0
8	**9270**	**208**	**2.24**
8	12	0	0
CO	3390	206	6.08
MT	642	0	0
ND	324	0	0
SD	642	2	0.31
UT	3774	0	0
WY	486	0	0
9	**51006**	**224**	**0.44**
9	354	3	0.85
AS	408	1	0.25
AZ	6648	47	0.71
CA	38808	133	0.34
GU	1236	19	1.54
HI	1920	0	0
MP	270	21	7.78
NN	138	0	0
NV	1224	0	0
10	**11286**	**15**	**0.13**
10	48	0	0

EPA Region	Number of Analytical Results	Number of Analytical Results > MRL	% > MRL
State/Territory			
AK	414	0	0
ID	2064	0	0
OR	2526	0	0
WA	6234	15	0.24
US Summary	**221831**	**1152**	**0.52**

Table A.4. UCMR3 results at entry points (EPs) sourced by surface water facilities where at least one PFAS concentration was reported in New York (ng L^{-1}).



Reported PFAS Concentrations in Drinking Water at Entry Points Sourced by Surface Water Facilities in New York (ng L^{-1})

PWS ID Facility ID PFAS	**2013**				**2014**				**2015**			
NY3503549	1	2	3	4	5	6	7	8	9	10	11	12
90001												
PFHpA				**21**	**17**	**17**	**19**					
PFHxS				**70**	**65**	**57**	**59**					
PFOA				**27**	**27**	**25**	**27**					
PFOS				**170**	**150**	**140**	**140**					
NY3503580												
31449												
PFHpA							**19**	**18**	▨	▨		
PFHxS							**48**	**54**	▨	▨		
PFOA							**29**	**22**	▨	▨		
PFOS							**140**	**128**	▨	▨		

▨ <MRL
PFHxS
PFHpA
PFOA
PFOS
PFNA

Table A.5. UCMR3 results at entry points (EPs) sourced by groundwater facilities where at least one PFAS concentration was reported in North Carolina (ng L^{-1}).

Reported PFAS Concentrations in Drinking Water at Entry Points Sourced by Groundwater in North Carolina (ng L^{-1})

PWS ID Facility ID	PFAS	**2013**				**2014**				**2015**		
		1	2	3	4	5	6	7	8	9	10	11
NC0326127												
FID-113	PFOA					**20**		▨				
FID-118	PFOA					**21**		▨				
NC0326332												
FID-30	PFOS						▨	**41**				
NC0363108												
FID-39	PFHxS	**42**		**40**								
	PFOS	**76**		**73**								
NC0392373												
FID-90003	PFHxS								**94**	**110**		
NC0464020												
FID-90004	PFOS									▨		**53**
NC0474045												
FID-90005	PFOS					**50**		▨				

▨ <MRL

PFHxS

PFHpA

PFOA

PFOS

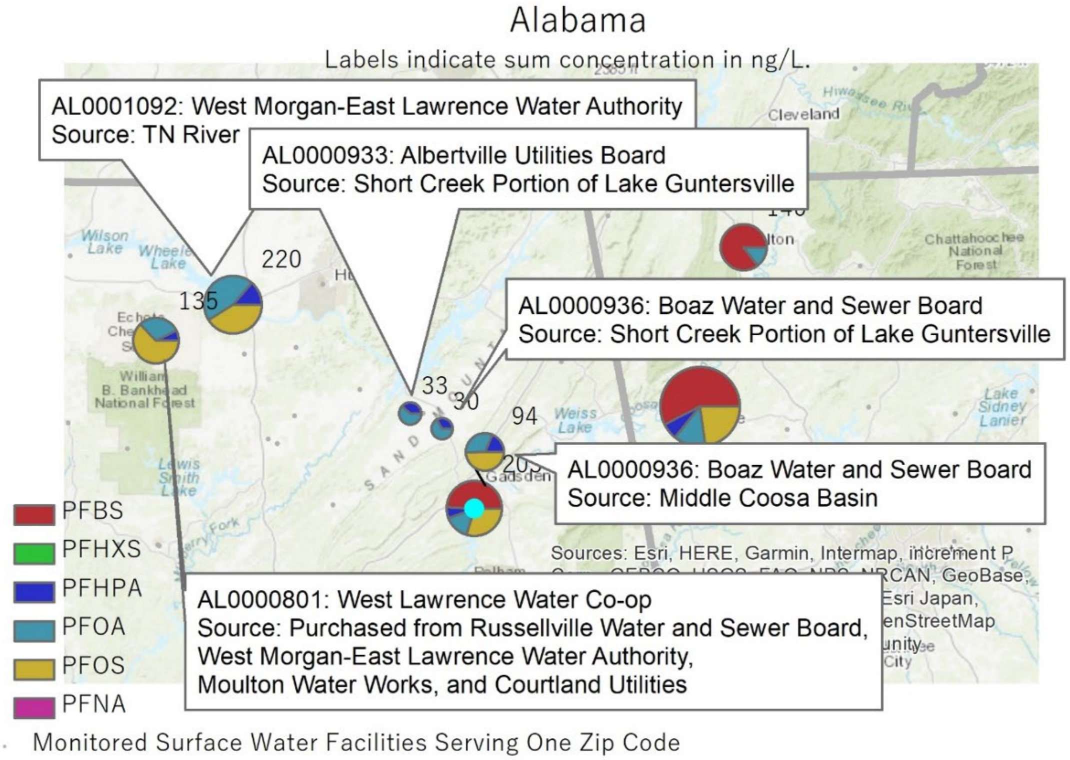
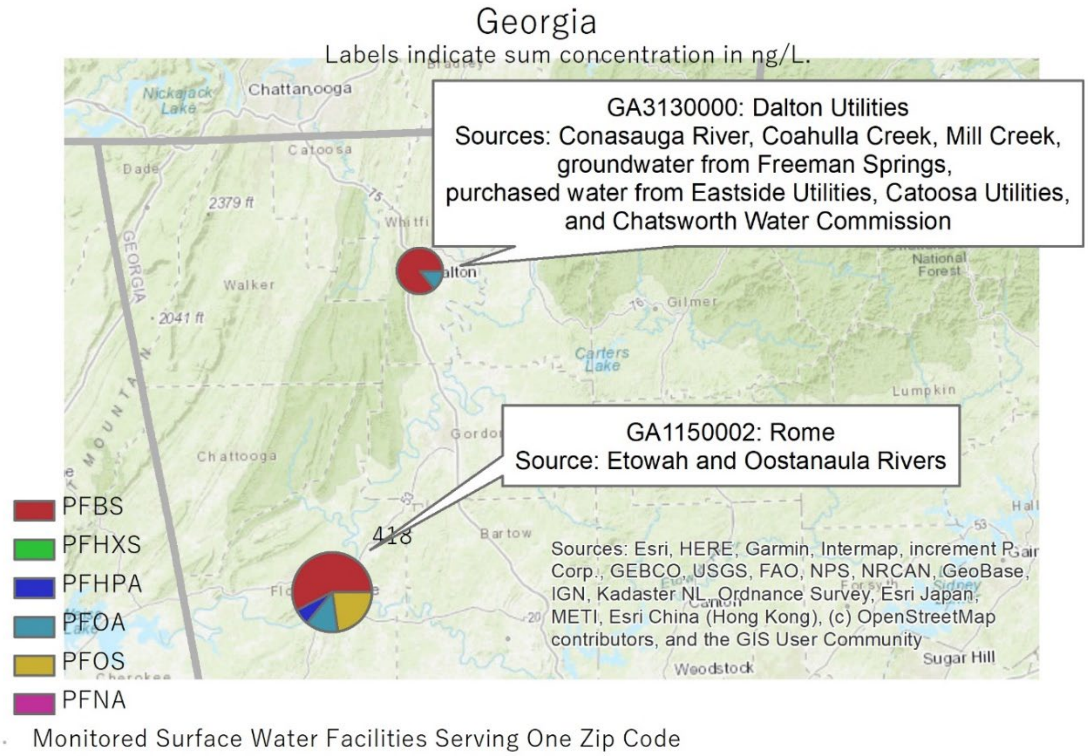


Figure A.1. Source water information from Consumer Confidence Reports (CCRs) for surface water systems where PFAS were reported in drinking water in North Alabama.



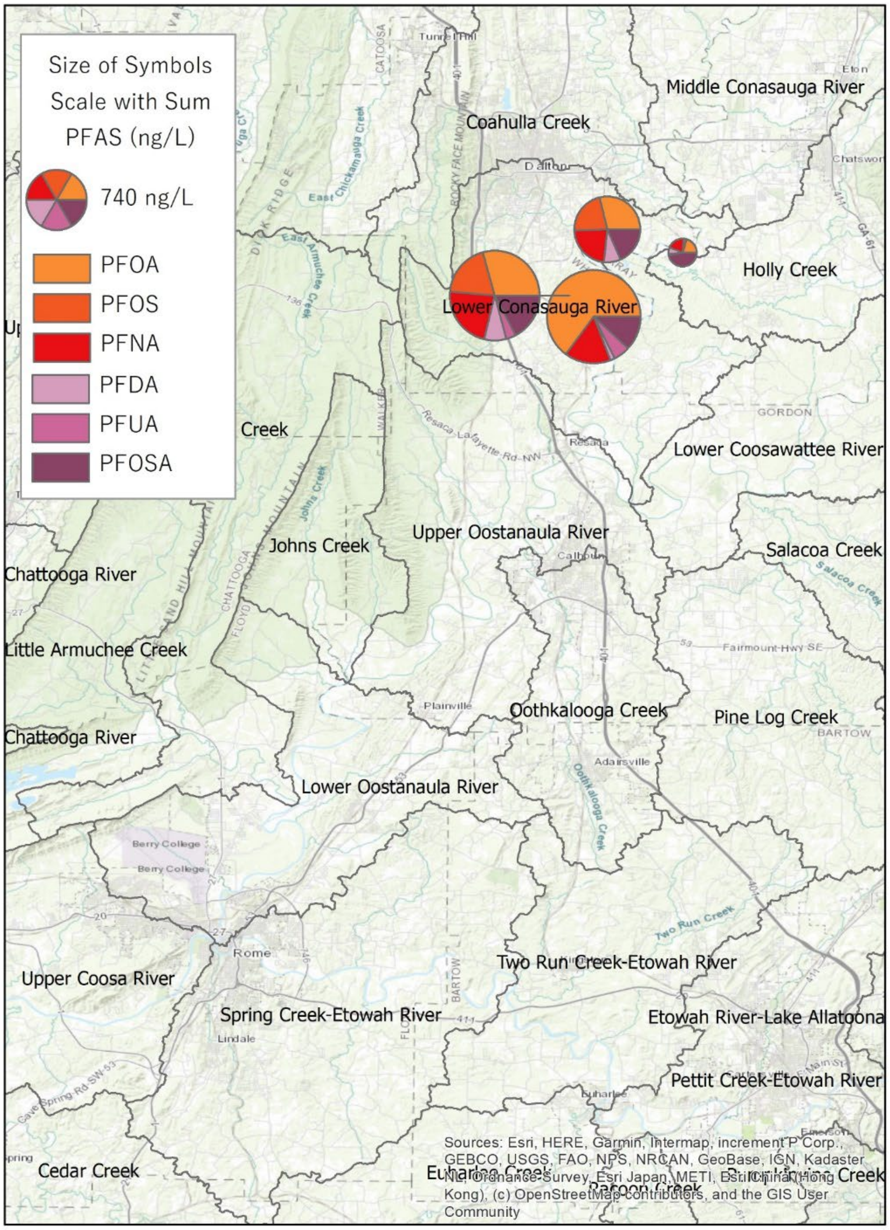
GA3130000: Dalton Utilities
Sources: Conasauga River, Coahulla Creek, Mill Creek, groundwater from Freeman Springs, purchased water from Eastside Utilities, Catoosa Utilities, and Chatsworth Water Commission

GA1150002: Rome
Source: Etowah and Oostanaula Rivers

Sources: Esri, HERE, Garmin, Intermap, increment P Corp., GEBCO, USGS, FAO, NPS, NRCAN, GeoBase, IGN, Kadaster NL, Ordnance Survey, Esri Japan, METI, Esri China (Hong Kong), (c) OpenStreetMap contributors, and the GIS User Community

Figure A.2. Source water information from Consumer Confidence Reports (CCRs) for surface water systems where PFAS were reported in drinking water in Northwest Georgia.

Konwick et al. 2008, Samples Collected March 2006



PFBA, PFBS, PFPeA, PFHxA, PFHxS, and PFHpA were not analyzed.

Figure A.3. Historic PFAS occurrence in Northwest Georgia surface water, 2006 (Konwick et al., 2008).

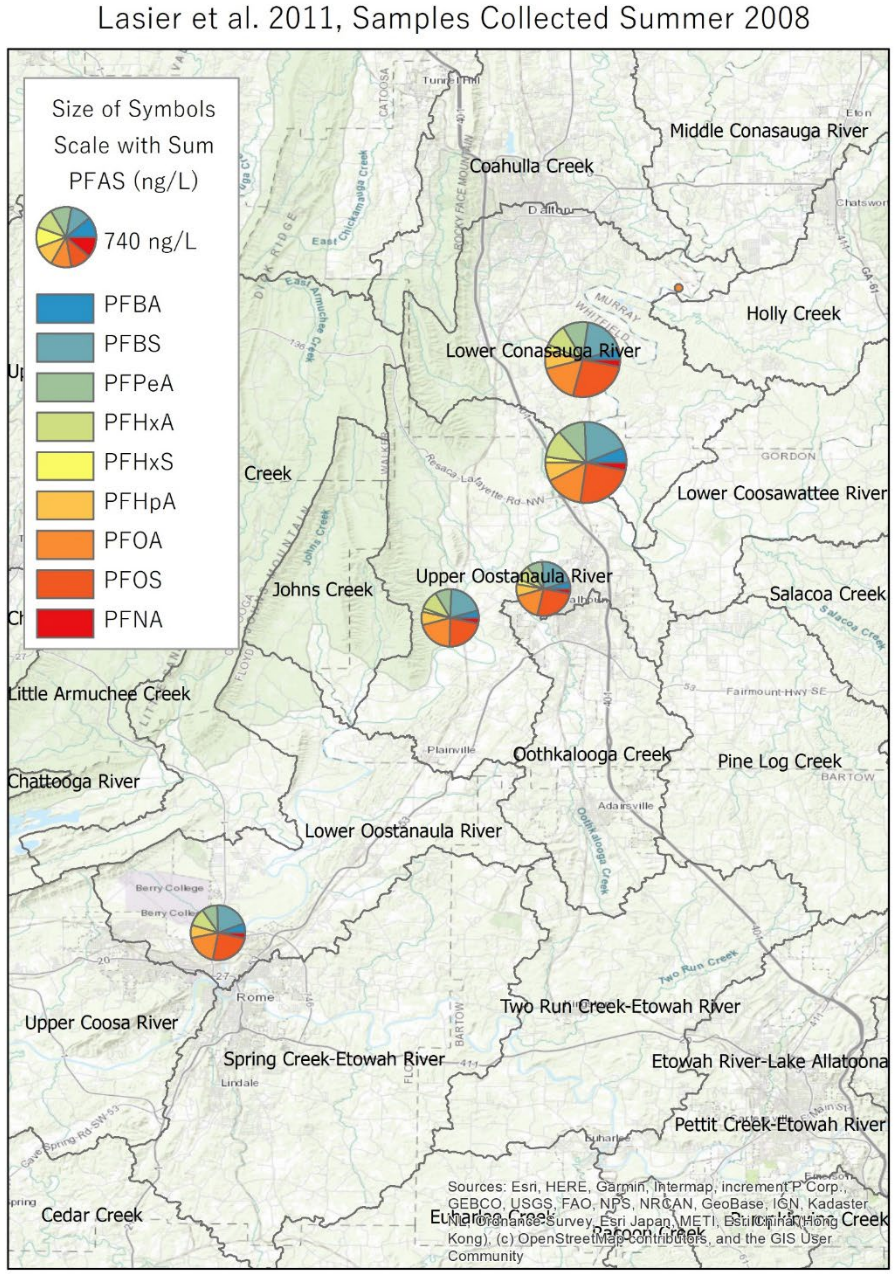
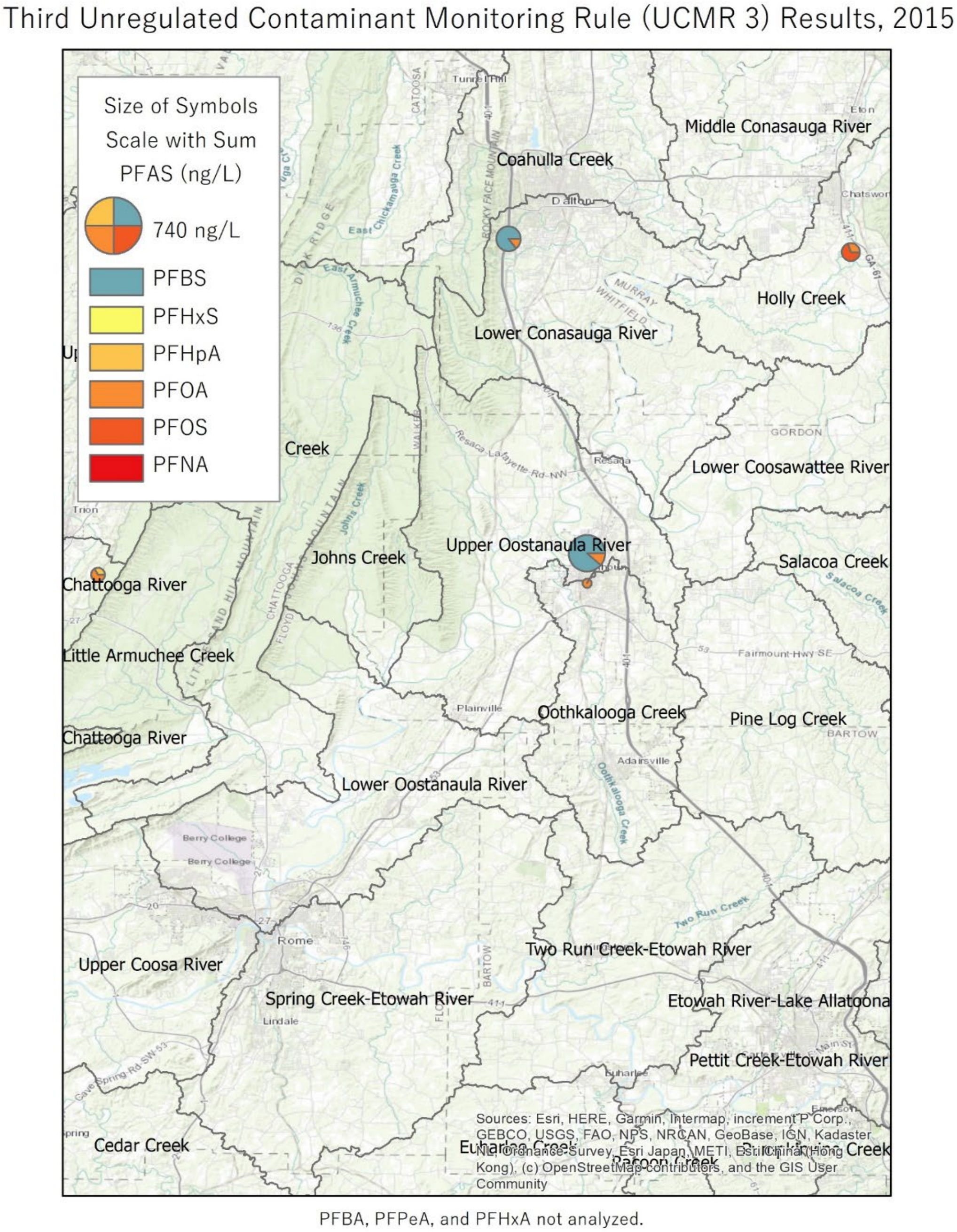


Figure A.4. PFAS occurrence in Northwest Georgia surface water, 2008 (Lasier et al., 2011).


Third Unregulated Contaminant Monitoring Rule (UCMR 3) Results, 2015

PFBA, PFPeA, and PFHxA not analyzed.

Figure A.5. PFAS occurrence in Northwest Georgia drinking water, 2015 (UCMR3).

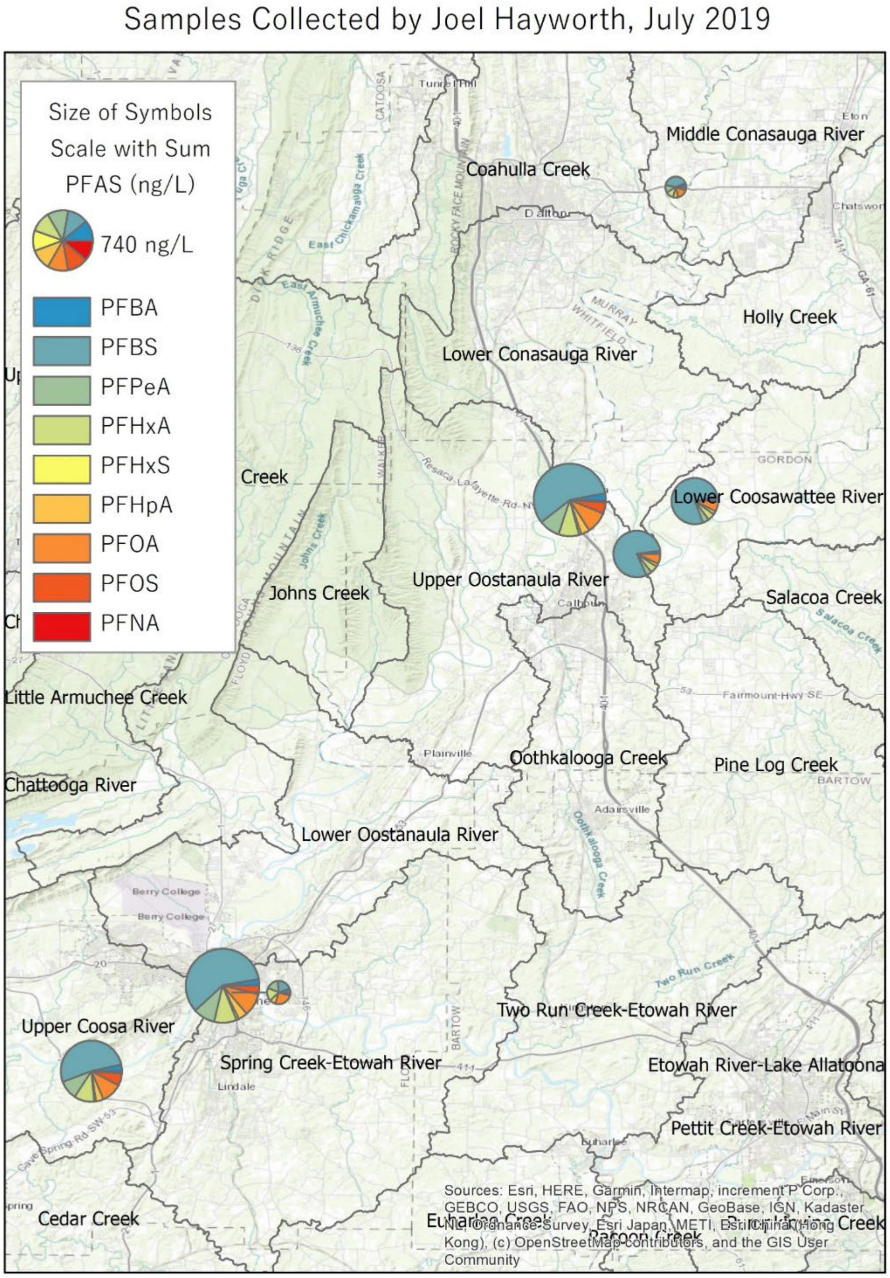
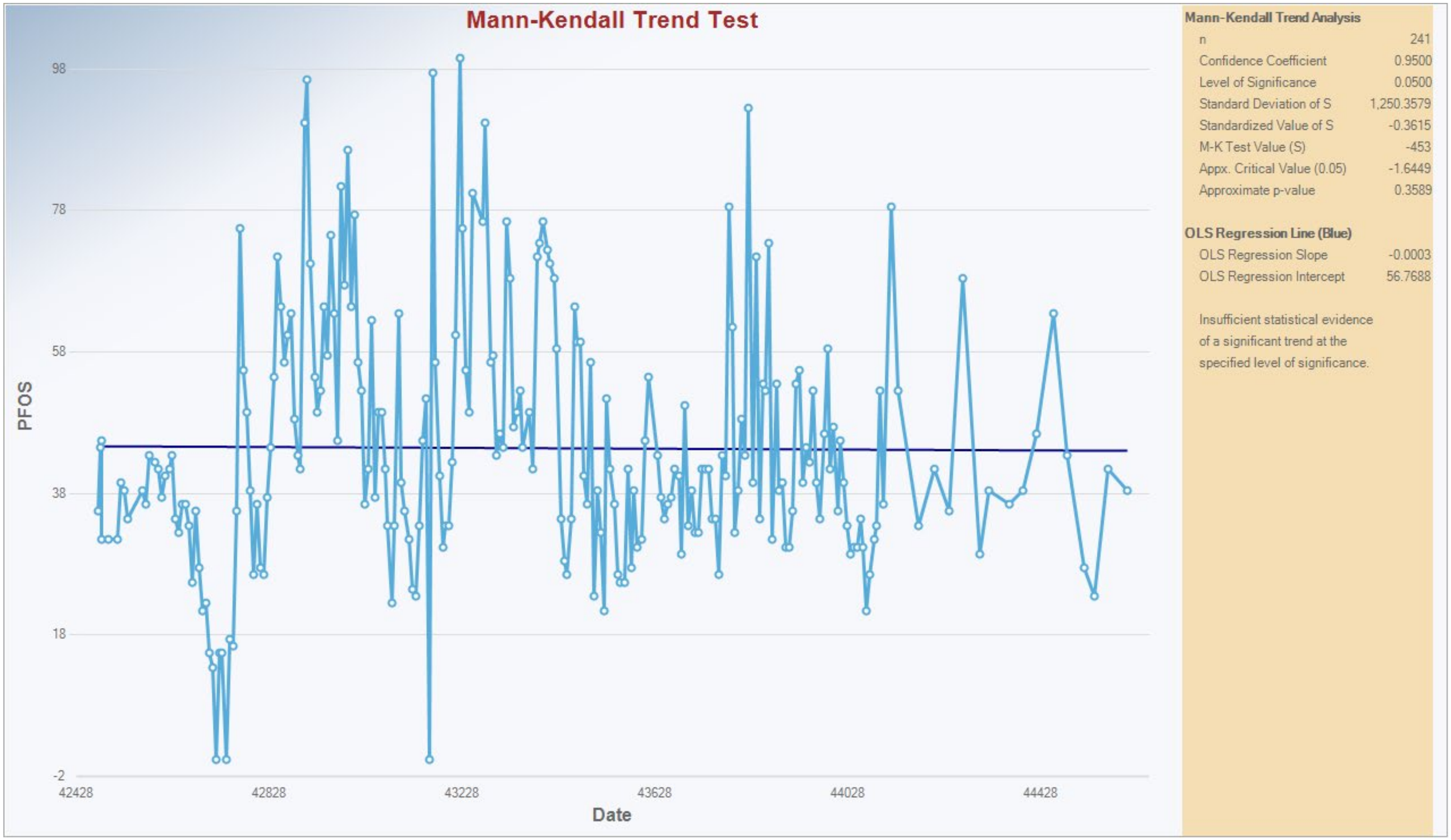


Figure A.6. PFAS occurrence in Northwest Georgia surface water, 2019.



Mann-Kendall Trend Analysis

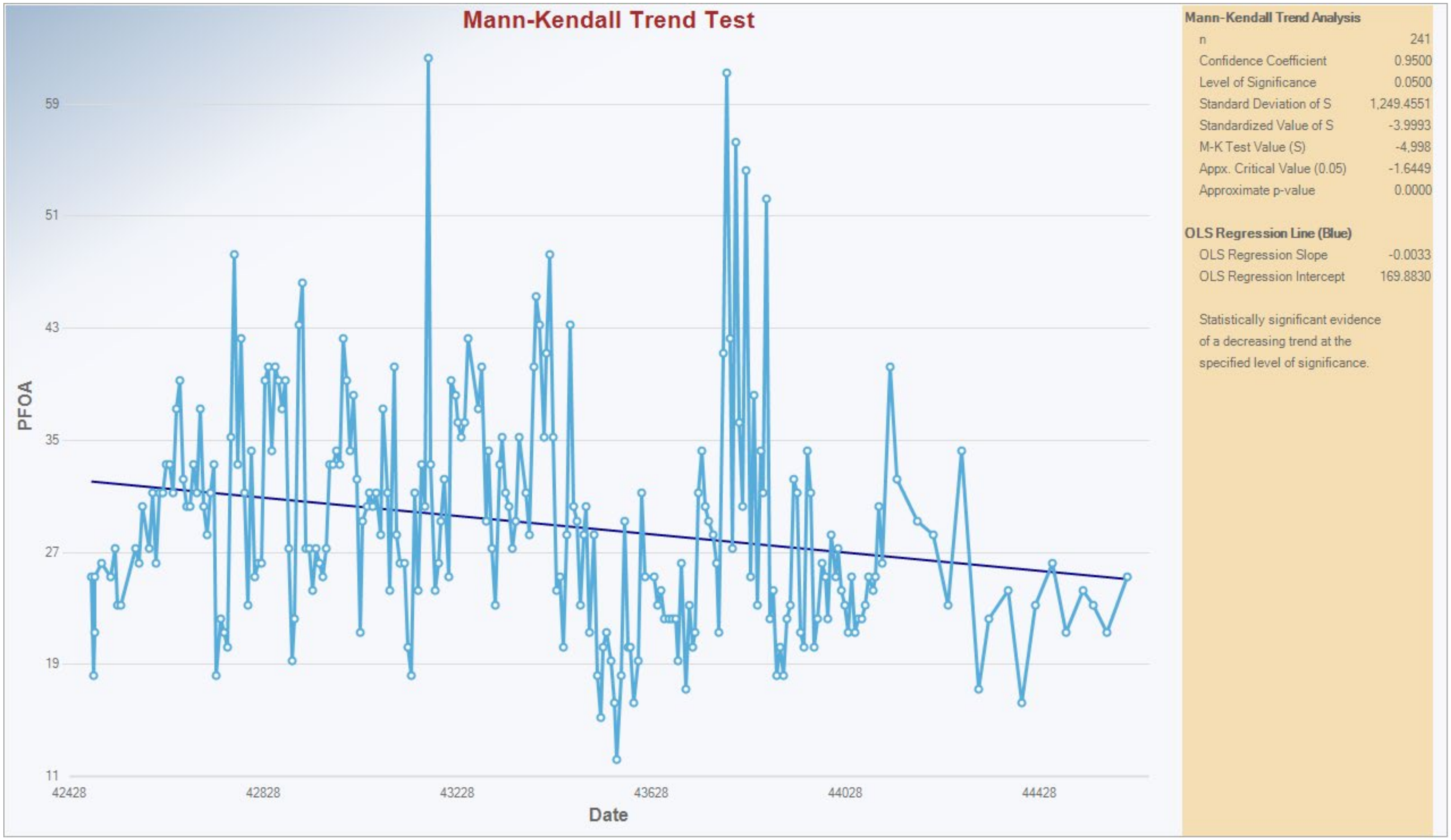
n	241
Confidence Coefficient	0.9500
Level of Significance	0.0500
Standard Deviation of S	1,250.3579
Standardized Value of S	-0.3615
M-K Test Value (S)	-453
Appx. Critical Value (0.05)	-1.6449
Approximate p-value	0.3589

OLS Regression Line (Blue)

OLS Regression Slope	-0.0003
OLS Regression Intercept	56.7688

Insufficient statistical evidence of a significant trend at the specified level of significance.

Figure A.7. Mann-Kendall trend test: PFOS concentration in raw water (Coosa River) at AL0000577 (April 2016 – February 2022)



Mann-Kendall Trend Analysis

n	241
Confidence Coefficient	0.9500
Level of Significance	0.0500
Standard Deviation of S	1,249.4551
Standardized Value of S	-3.9993
M-K Test Value (S)	-4,998
Appx. Critical Value (0.05)	-1.6449
Approximate p-value	0.0000

OLS Regression Line (Blue)

OLS Regression Slope	-0.0033
OLS Regression Intercept	169.8830

Statistically significant evidence of a decreasing trend at the specified level of significance.

Figure A.8. Mann-Kendall trend test: PFOA concentration in raw water (Coosa River) at AL0000577 (April 2016 – February 2022)

Note: The data used in the analyses shown in Figures A.7 and A.8 above were acquired using the Alabama Department of Environmental Management (ADEM) eFiles search engine. Per the instructions provided on ADEM’s “Per- and Polyfluoroalkyl Substances (PFAS) in Drinking Water” webpage (ADEM, 2022b)*, the PWSID of interest (AL0000577) was entered into the “Permit Number” field, “PFC” was entered in the “File Name” field, and “Water” was selected as the “Media Area.” The file that contained the following data can be found by directly searching for the File Name “AL0000577 01055 02-25-2022 DWMORG WEB GADSDEN PFC LAB DATA” with “Water” selected as the “Media Area.”*