# EVALUATION OF DRINKING WATER CONTAMINANTS IN A PERI-URBAN NEIGHBORHOOD AFTER CONNECTION TO MUNICIPAL WATER SERVICE

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## ABSTRACT

April Desclos: Evaluation of Drinking Water Contaminants in a Peri-Urban Neighborhood After Connection to Municipal Water Service (Under the direction of Jacqueline MacDonald Gibson)

In a practice called "municipal underbounding", many African American communities in the southern United States have been excluded from town boundaries and must rely on private wells that have worse water quality than community water systems. In this study, I collected water samples from 12 households in a majority African American, underbounded community in North Carolina to assess the change in water quality after households connected to municipal water. I compared levels of metals and per- and polyfluoroalkyl substances (PFAS) in private well and municipal water samples and used samples collected sequentially to determine whether running the faucet (flushing) reduces exposure to lead. After connection, levels of lead and copper decreased significantly, and lead levels remained low. Flushing for 15 seconds significantly reduced lead levels for both water sources. The average, total PFAS concentration was higher in Apex water than in private wells, but this difference was not significant. This thesis is dedicated to my husband, Russell, and my daughter, Autumn. Thank you for loving and supporting me and helping me stay true to myself.

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# LIST OF ABBREVIATIONS

AAP	American Academy of Pediatrics
AI	Aggressive index
CSMR	Chloride-to-sulfate mass ratio
EPA	Environmental Protection Agency
ETJ	Extraterritorial jurisdiction
HDPE	High density polyethylene
ICP-MS	Inductively coupled plasma mass spectrometry
ICP-OES	Inductively coupled plasma optical emission spectroscopy
LMER	Linear mixed effects regression
LSI	Langelier saturation index
LSK	Larson Skold index
MCL	Maximum contaminant level
MDL	Method detection limit
MRL	Method reporting limit
PFAS	Per- and polyfluoroalkyl substances
PFECA	Perfluoroether carboxylic acid
PFESA	Perfluoroether sulfonic acid
PFCA	Perfluoroalkyl carboxylic acid
PFSA	Perfluoroalkane sulfonic acid
SWDA	Safe Drinking Water Act
UNC	University of North Carolina

#### **1. INTRODUCTION**

In the southern United States, the boundaries of many small cities and towns have been drawn to exclude African Americans and leave them on the fringes of towns, a practice called "municipal underbounding" (Aiken, 1987; Johnson et al., 2004; Marsh et al., 2010; Parnell et al., 2004). In North Carolina, municipalities can legally underbound communities by creating extraterritorial jurisdictions (ETJs) that extend one to three miles past the town's limits (N.C. Gen. Stat. §160D-202(a)). Within these areas, towns can exercise planning and zoning authority, but residents cannot vote in local elections and do not have access to basic municipal services such as water and sewer. Residents can petition for voluntary annexation, bringing their community inside the town's boundaries. However, residents bear the cost of annexation and connection to water and sewer service unless they can obtain public financing or other support (Marsh et al., 2010; Wilson et al., 2008). Towns are less likely to annex African American communities compared to white communities, indicating that race can play a role in annexation decisions (Aiken, 1987; Johnson et al., 2004; Lichter et al., 2007).

Racial disparities in access to municipal water service exist for underbounded communities in North Carolina (Marsh et al., 2010; Parnell et al., 2004). In Wake County, NC, a 10% increase in the proportion of African Americans living in ETJs results in 3.8% higher odds of exclusion from water service (Gibson et al., 2014). Public water systems must comply with enforceable water quality standards called maximum contaminant levels (MCLs), including standards to prevent health impacts (U.S. EPA, 1996). Additionally, if more than 10% of system-wide samples exceed the "action level" of 15 µg/L for lead or 1,300 µg/L for copper,

mitigation efforts such as source water treatment or lead service line replacement must be made (Control of Lead and Copper, 1991). However, MCLs and requirements for lead and copper monitoring do not apply to systems with fewer than 15 service connections or that serve fewer than 25 individuals (U.S. EPA, 1996), including private wells. Testing for contaminants and treating water is the private well owner's responsibility (Fox et al., 2016; Zheng & Flanagan, 2017).

Previous studies have demonstrated a higher risk of contamination for private wells in ETJs compared to non-ETJ households with municipal water. One study found that the proportion of households in Wake County ETJs with private wells exceeding the action level for lead was 16 times as high as nearby households with municipal water and more than four times as high as Wake County households that rely on private wells and do not live in ETJs. 10% of samples exceeded the action level for copper (Stillo and Gibson, 2018). In a majority African American, underbounded community located near a landfill in Orange County, NC, the pH of most private wells was lower than the recommended 6.5, putting them at risk for elevated metals from corrosion. 42% of wells exceeded the MCL for iron and manganese, and 8% exceeded the action level for lead (Heaney et al., 2013). Underbounded communities relying on private wells in North Carolina have also experienced a higher prevalence of fecal coliforms (Heaney et al., 2013; Heaney et al., 2011) and *E. coli* (Stillo and Gibson, 2016) compared to nearby communities on municipal water.

Extending municipal water service could prevent many health impacts associated with contaminants in private wells, including 22% of annual emergency department visits for acute gastrointestinal illness in Wake Count ETJs (Stillo and Gibson, 2016). Wake County children relying on private wells have 25% increased odds of elevated blood lead compared to children on

regulated, community water systems, and children living in ETJs in Wake County have significantly higher blood lead than children living within town boundaries or in rural areas (Gibson et al., 2020). Numerous health effects associated with lead have been documented in children, including intellectual disabilities (Delgado et al., 2018; Miranda et al., 2007; Sorensen et al., 2018), developmental delays (Delgado et al., 2018), and attention deficit and hyperactivity (Goodlad et al., 2013; Lin et al., 2019). In adults, lead is associated with cardiovascular disease (Lanphear et al., 2018) and decreased kidney function (Harari et al., 2018). Although copper is an essential micronutrient, elevated levels in water can cause gastrointestinal illness (Dietrich et al., 2004).

Elevated lead and copper levels in private wells are typically caused by corrosion of premise plumbing (Mulhern and Gibson, 2020; Pieper et al., 2018a; Pieper et al., 2015b; Stillo and Gibson, 2018) or well components (Pieper et al., 2018a; Pieper et al., 2015b). Factors contributing to corrosivity of water include low pH (Schock, 1989) and a high chloride to sulfate mass ratio (CSMR) (Nguyen et al., 2011; Pieper et al., 2018b). Public water systems take measures to reduce corrosivity, including increasing the pH and dosing water with corrosion control chemicals that form a protective coating on pipes (Bae et al., 2020; Pieper et al., 2018b).

Another group of chemicals of concern in drinking water are per- and polyfluoroalkyl substances (PFAS), which include more than 9,000 individual species (U.S. EPA, 2020). PFAS are a group of highly fluorinated, aliphatic chemicals with at least one carbon (Buck et al., 2011) and strong carbon-fluorine bonds that make them high stabile and persistent in the environment (Bentel et al., 2019). They have been manufactured and used for many commercial and industrial applications in the United States since the 1940s (Glüge et al., 2020). Their dual hydrophobic and hydrophilic nature makes them useful (Buck et al., 2011) as a surfactant,

friction reducer, and repellent of water and grease (Kwiatkowski et al., 2020). More than 200 use and subuse categories have been identified for more than 1,400 PFAS species as varied as firefighting foam, electronic devices, dental floss, waterproof clothing, artificial grass, and food packaging (Glüge et al., 2020). Since they are highly mobile, PFAS can travel long distances from their source and are globally distributed in the environment (Gomis et al., 2015; Yeung et al., 2017). They are bioaccumulative (Conder et al., 2018) and are associated with a variety of health outcomes, including kidney and testicular cancer (Barry et al., 2013; Vieira et al., 2013), thyroid disorders (Lopez-Espinoza et al., 2012; Blake et al., 2018), decreased kidney function (Blake et al., 2018), ulcerative colitis (Steenland et al. 2013), pregnancy-induced hypertension (Darrow et al., 2013), reduced immune response to vaccinations (Grandjean et al., 2012; Granum et al., 2013; Looker et al., 2014; Stein et al., 2016), and high cholesterol (Frisbee et al., 2010; Nelson et al., 2010).

In North Carolina, the occurrence of PFAS in surface waters and community water supplies has been extensively studied, showing widespread occurrence (Herkert et al., 2020; Kotlarz et al., 2019; Saleeby et al., 2021; Strynar et al., 2015; Sun et al., 2016). Information on PFAS occurrence in private wells is more limited. PFAS have been detected in wells near a PFAS manufacturing plant in the Cape Fear River watershed (Hopkins et al., 2018) and in central North Carolina (Herkert et al., 2020). They can enter wells from groundwater contamination by industrial or agricultural sources or from septic system wastewater containing PFAS from cookware, food packaging, clothing, or other products (Schaider et al., 2016). No information is available specifically on the occurrence of PFAS in private wells in underbounded communities.

Evidence to date suggests that underbounded communities in North Carolina experience worse drinking water quality than neighboring areas served by regulated water systems. However, the effects of extending water service to an underbounded community have not been studied previously. In this thesis, I take advantage of a natural experiment to assess the effect on drinking water quality in an underbounded neighborhood following its connection to municipal water. Specifically, I ask three questions:

1. What are the effects of switching from private well water to a community water system on concentrations of lead and other metals at the kitchen tap?

2. How does running the tap, or flushing, affect exposure to lead in drinking water in underbounded communities, and how does this change after switching to a community water system?

3. What are the effects of switching from private well water to a community water system on the types and concentrations of PFAS at the kitchen tap?

#### 2. DESCRIPTION OF STUDY COMMUNITY AND MUNICIPAL WATER SYSTEM

Irongate Drive is a neighborhood of 24 households located in Apex, North Carolina (Figure 1). In contrast with Apex, which is 79.5% Caucasian and has a median household income of \$105,404 (U.S. Census Bureau, 2019 [race]; 2015-2019 [income]), Irongate Drive is 78.6% African American, with a median income of less than \$60,000 (Lockhart et al., 2020). Before 2020, Irongate Drive was located within an ETJ and bordered Apex on two sides. Although a municipal water line ended at the neighborhood's entrance, it had no municipal water or sewer service and relied on private wells and septic systems (Lockhart et al., 2020). Water insecurity was demonstrated by a high percentage of wells (80%) failing to provide enough water and fear of not having enough water in the future. When wells ran dry, residents could not perform necessary tasks such as bathing and cooking and were forced to rely on bottled water (Lockhart et al., 2020), concerns that were heightened during the COVID-19 pandemic when water use increased due to family members working from home. Well maintenance costs were also high, averaging about \$1,400 per year and as high as \$20,000 per year (Lockhart et al., 2020).

Irongate residents spent many years trying to annex to the Town of Apex and receive town services. With the help of stories about water shortages in the neighborhood, research on well contamination, and assistance from human rights attorneys and the town manager, the neighborhood's petition for annexation recently succeeded. On January 21, 2020, Apex annexed Irongate Drive, bringing it into the town's corporate limits and making it eligible to receive

municipal services (Apex, NC, 2020). Water mains were installed in the neighborhood during March 2020, and households began connecting to municipal water service during May.



Figure 1: Irongate community with new water main (modified from Lockhart et al., 2020)

Apex municipal water is sourced from the B. Everett Jordan Lake and is treated at a plant jointly owned by the Town of Apex and Town of Cary (Town of Apex, 2019). Water treatment includes conventional sedimentation and filtration, ozonation, and powdered activated carbon to reduce levels of PFAS (Monschein, 2020). The water is disinfected with chloramines, except during March when only chlorine is used (Town of Cary, n.d.). It is also dosed with orthophosphates to prevent corrosion of water pipes (Town of Cary, 2019). In addition to testing metals for compliance with MCLs, the Cary/Apex Water Treatment Plant conducts quarterly testing of 39 PFAS compounds (Monschein, 2020). The Town of Apex owns and maintains the town's water distribution system. Most of the system's water mains (93.6%) are ductile iron, and the remaining water mains are made of cast iron, polyvinyl chloride, or asbestos cement (NC DEQ, 2019). The new water mains installed in the Irongate Drive neighborhood are made of ductile iron (Town of Apex, n.d.). For significant dead ends of the distribution system, such as Irongate Drive, the town regularly flushes hydrants. Flushing involves opening hydrants and releasing water to the street at the lowest flow rate necessary to maintain adequate disinfectant residual and water quality. During the study, flushing occurred continuously from approximately July 22<sup>nd</sup> through July 30<sup>th</sup>, once on July 31<sup>st</sup> and August 26<sup>th</sup>, every three hours for 50 minutes at a time from August 26<sup>th</sup> through September 21<sup>st</sup>, and continuously from September 21<sup>st</sup> through the remainder of the study.

#### **3. MATERIALS AND METHODS**

## 3.1 Participant Recruitment

Participants were initially recruited from the 21 households who participated in a survey as part of a previous study of water quantity issues at Irongate Drive (Lockhart et al., 2020). These households received a mailed letter and a water test kit at their doorstep that included collection bottles, sampling instructions, and a household survey. The 12 households who participated in the first sampling event received a second letter inviting them to participate in the remaining scheduled events and a second water test kit at their doorstep. Three of these 12 households dropped out of the study after the first sampling event, six households participated in all remaining sampling events, and three households participated in most events (see Table 1 for the number of households that participated in each event). Participation was completely voluntary, and households received a gift card as compensation after each sampling event.

#### 3.2 Household Survey

Each water test kit on all sampling dates contained a household survey (Appendix 1). Surveys included questions about the household's private well, including depth and age, and the household's septic system, including age and previous and current problems. Questions about household plumbing were also covered, including water pipe material, water treatment system, maintenance or changes to plumbing or treatment systems between sampling events, information to calculate the kitchen faucet's flow rate, and whether an aerator was installed on the kitchen faucet. Home age and date of connection to Apex water service were also requested.

## 3.3 Sample Collection

Two days before each sampling event, participants received a test kit with new, widemouth high density polyethylene (HDPE) bottles that were either certified for metals analysis or soaked with 3 M nitric acid for at least three days and then rinsed five times with deionized water. Participants were asked to collect samples at their kitchen faucet after at least six hours of stagnation and before household members used any water. Samples that were known to be collected after water was used (i.e., not first draw) were noted. Participants were not asked to remove or bypass faucet aerators or water treatment systems that were in place during sample collection. Sample bottles were collected from participants' porches on the morning of sample collection and transported to the University of North Carolina (UNC).

Sampling included two methods, first-draw sampling and sequential sampling, which are described below. Table 1 shows the method and number of households connected to private wells or municipal water for each sampling date. Five of the 12 participating households connected to municipal water during the study period: three households on May 20<sup>th</sup> and two households on July 6<sup>th</sup>.

Sampling Date	Method	Participant Water Source (number of households)			
		Private Well	Municipal Water		
3/23/2020	Sequential sampling	12	0		
6/29/2020	First draw	5	3		
7/13/2020	First draw	4	5		
7/27/2020	Sequential sampling	4	4		
8/10/2020	First draw	4	3		
8/10/2020	Sequential sampling	0	1		
8/24/2020	First draw	4	4		
9/14/2020	First draw	4	5		
9/28/2020	Sequential sampling	4	3		
10/20/2020	Sequential sampling	0	2		

**Table 1.** Sampling design and participant water source

#### 3.3.1 First-Draw Sampling

First, participants turned on their kitchen faucet, immediately collected one 250 mL sample (bottle 1), turned off their faucet, and closed the bottle lid. They immediately repeated these steps for one 1 L sample (bottle 2). The 250 mL bottle was used to isolate contaminants contributed by the faucet and first several feet of premise plumbing. Pieper et al. (2015b) estimated that the volume of water in the first eight feet of plumbing from the faucet is 250 mL. *3.3.2 Sequential Sampling* 

It can be difficult to identify the source of metal contaminants in drinking water because plumbing configurations cannot always be easily determined by inspecting homes or reviewing records (Lytle et al., 2019). The standard sampling procedure under the Lead and Copper Rule is to take a first-draw, 1-L sample (40 C.F.R. § 141.86), which only estimates levels of metals from the faucet and first few feet of plumbing. Instead, sequential sampling or "lead profiling" can be used, which generally involves surveying plumbing lengths and diameters and visible fixtures, collecting successive samples, and relating sample volume to plumbing volume to identify sources from the faucet to the private well or to the water main for homes on municipal water. Correlations between lead and other metals can help to identify specific plumbing materials contributing lead to water (Lytle et al., 2019). In addition to targeting plumbing components for removal, sequential sampling can help determine whether corrosion control is effective throughout the plumbing system and measure the risk of lead exposure from running the faucet, or flushing. Flushing can effectively reduce lead exposure in some homes, but it can also increase exposure if particulate lead is dislodged from pipes or if distant plumbing sources are leaching lead (Lytle et al., 2019). The effectiveness of flushing depends on factors such as plumbing configuration, whether lead is in the dissolved or particulate form, water use patterns,

and disturbances of plumbing or service lines (Katner et al., 2018; Pieper et al., 2015b). Previous studies have used sequential sampling to identify sources of lead and the effectiveness of flushing in homes with municipal water (Deshommes et al., 2018; Deshommes et al., 2017; Katner et al., 2018; Lytle et al., 2019; Triantafyllidou et al., 2015; Trueman et al., 2016) and in homes relying on private wells (Mulhern and Gibson, 2020; Pieper et al., 2018a; Pieper et al., 2015b).

Participants followed the sequential sampling method described by Mulhern and Gibson (2020). In summary, participants collected one 250 mL sample (bottle 1) followed immediately by three 1 L samples (bottles 2-4), ran the water at full flow for one minute, collected a fourth 1 L sample (bottle 5), ran the water at full flow for five more minutes, and then collected a fifth 1 L sample (bottle 6). For homes with private wells, the first four samples (1-3.25 L) are estimated to come from the length of plumbing from the faucet to the pressure tank (Pieper et al., 2015b), while the fifth and sixth samples are estimated to come from the pressure tank and borehole (Mulhern and Gibson, 2020). Katner et al. (2018) estimated that it takes approximately 2.2 to 3.0 minutes of flushing to purge the water in premise plumbing and service lines, assuming a typical pipe diameter of ¾ inches and a faucet flow rate of 3.0 L per minute. Based on these estimates and an average flow rate of 4.6 L per minute for Irongate homes without faucet aerators, it is assumed that the first four Apex water samples are from the premise plumbing, the fifth sample is from the service line or water main, and the sixth sample is from the water main. *3.4 Sample Filtration and Preservation* 

On the day of sample collection, 10 mL aliquots were pipetted from all samples for analysis of metals. To quantify dissolved lead, a second set of 10 mL aliquots were filtered

through 0.45  $\mu$ m, GE Whatman GD/XP syringe filters. Filtered and unfiltered aliquots were acidified to 2% nitric acid, stored at 4° C, and digested for at least 16 hours before analysis. *3.5 Water Quality Analyses* 

#### 3.5.1 Metals Analysis

On the day of sample collection, basic water quality parameters were measured at UNC. Conductivity and pH were measured with a portable meter (HI98129) following a two-point calibration. Phenolphthalein and total alkalinity were measured with a Hach test kit (AL-AP). Conductivity, pH, and alkalinity were tested on the fifth sample bottle on March 23<sup>rd</sup>, September 28<sup>th</sup>, and October 20<sup>th</sup>, and on the second sample bottle on all other dates.

The acidified, 10 mL aliquots were analyzed for lead (Pb), copper (Cu), beryllium (Be), aluminum (Al), vanadium (V), chromium (Cr), manganese (Mn), arsenic (As), iron (Fe), cobalt (Co), nickel (Ni), zinc (Zn), silver (Ag), cadmium (Cd), and antimony (Sb) using inductively coupled plasma mass spectrometry (ICP-MS) per EPA Method 200.8 (U.S. EPA, 1994). Method reporting limits (MRLs) were 0.2 µg/L for Pb, 0.5 µg/L for Al, and 0.1 µg/L for all other metals. QA/QC measures included instrument performance verification every 10 samples with solvent check solution and solvent blank solution, and processing of method blanks (27 total) and spiked method blank (54 total). All analytes were below the MRL in method blanks, and recovery was 91-108% for all analytes in spiked method blanks.

#### 3.5.2 Ions Analysis

For calculation of measures of corrosion and scaling potential, ions were tested on the fifth bottle for samples collected on September 28<sup>th</sup> and October 20<sup>th</sup>. Chloride and sulfate were analyzed on an ICS 3000 (Thermo Fisher, Waltham, MA) ion chromatograph. QA/QC measures included one duplicate sample, one spiked sample, method blank samples, spiked solvent

samples, and instrument performance verification every ten samples (solvent check solution and solvent blank solution). Calcium and magnesium were measured by inductively coupled plasma optical emission spectroscopy (ICP-OES) using an iCAP 7000 (Thermo Scientific) ICP-OES equipped with an ASX-520 autosampler (CETAC, Omaha, NB). QA/QC measures for calcium and magnesium also included instrument performance verification every ten samples.

#### 3.5.3 PFAS Analysis

PFAS were tested for all participants on March 23<sup>rd</sup> and only for participants connected to Apex water service on July 27<sup>th</sup>, August 10<sup>th</sup>, September 28<sup>th</sup>, and October 20<sup>th</sup>. The sixth sample bottle in the sequential sampling method was used for analysis to measure PFAS from the water source instead of from water within the premise plumbing. Levels of 25 PFAS compounds were measured using a method adapted from EPA Method 537 (Shoemaker et al., 2008). A positive and negative blank were run along with the samples. All analytes in the negative blank were <MRL and the average percent recovery for the positive blank (deionized water spiked with a known quantity of each analyte) was 98.6%.

# 3.6 Calculation of Measures of Corrosivity and Scaling Potential

Three measures of corrosivity were calculated: the aggressive index (AI), the chloride to sulfate mass ratio (CSMR), and the Larson-Skold index (LSK). The equation for AI (Ahmed et al., 2021) is:

$$AI = pH + \log(AH)$$

where A is total alkalinity (mg/L as CaCO<sub>3</sub>) and H is calcium hardness (mg/L as CaCO<sub>3</sub>). The equation for LSK (Masten et al., 2016) is:

$$LSK = ([Cl-] + [SO_42-])/(HCO_3- + [CO_32-])$$

where concentrations are in equivalents per liter. The Langelier saturation index (LSI) was also calculated to measure scaling potential. The equation for LSI (Masten et al., 2016) is:

$$LSI = pH - pH_{s} = pH + log \frac{[Ca^{2+}]\gamma_{Ca^{2+}}K_{a}[HCO_{3}^{-}]\gamma_{HCO_{3}^{-}}}{K_{sp}\gamma_{H^{+}}}$$

where  $\gamma$  is the activity coefficient,  $K_a$  is the acid dissociation constant for bicarbonate, and  $K_{sp}$  is the solubility product for calcium carbonate.

#### 3.7 Statistical Analysis

A regression modeling approach was used to determine the association between the dependent variable, concentrations of total PFAS (sum of all species) or metals in water, and water source (private wells or Apex water). For PFAS and for metals with values above the MRL in all samples, a linear mixed effect regression (LMER) approach was used to account for repeated sampling. If an individual PFAS chemical was detected in at least one sample for a water source, concentrations <MRL for that chemical and water source were replaced with MRL/ $\sqrt{2}$ . Analysis was performed in RStudio version 1.2.1335 using the lme4 package (Bates et al., 2015). To avoid biasing results by substituting values below the MRL (censored data), a Tobit approach (Tobin, 1958) for panel data was used to analyze metals with censored values. Analysis for these metals was performed with the censReg package (Henningsen, 2017). The distribution of metals concentrations was left-skewed, so the data were log-transformed. The regression equation is:

$$y_{it} = x'_{it}\beta + \mu_i + \epsilon_{it}$$

where  $y_{it}$  is the log of the metal concentration in household *i* at time *t*,  $\beta$  represents fixed effects (household water source),  $\mu_i$  represents a random intercept for each household, and  $\in_{it}$  is an error term. Reported *p* values in this thesis are from regression analyses, unless otherwise noted.

To find correlations between lead and other metals, the Spearman's test was used because data were non-normally distributed (Shapiro-Wilk test, p < 0.001).

#### 4. RESULTS AND DISCUSSION FOR METALS

#### 4.1 Household Characteristics

Table 2 lists home age and plumbing and septic system characteristics reported by participants on household surveys. Five homes (42%) were built before 1986, when the Safe Drinking Water Act (SWDA) was amended to limit lead in solder and flux to 0.2% and lead in pipes to 8% in public water systems or facilities providing drinking water (Safe Drinking Water Act Amendments of 1986). Six homes (50%) were built between 1986 and 2014, when the SWDA was amended again to limit lead in pipes, fittings, and fixtures to 0.25% (Reduction of Lead in Drinking Water Act). Most wells (70%) were older than 25 years. These wells are more likely to have lead in "packers" that help seal wells and to have submersible pumps with leadedbrass or other leaded well components (CDC, 2015; Pieper et al., 2018a). All reported well depths were less than 600 feet, and 5 wells (63%) were 150 to 200 feet deep. Shallower wells may be more vulnerable to contamination from septic systems and other surface sources compared to deeper wells, but this difference may be unimportant for drilled wells within the range of depths reported by Irongate residents (Fram and Belitz, 2011; Richards et al., 1996; Schaider et al., 2016). In five homes (45%), sediment filters, which can remove manganese, iron and some other contaminants attached to particles (Dvorak & Skipton, 2008), were in place during the study. One home had a carbon filter, which can effectively remove metals and PFAS (Mulhern and Gibson, 2020; Purchase et al., 2020). The majority (80%) of household water pipes were made of plastic. Most septic systems (70%) were 20 years or older, and 30% of households reported a problem with either their own or a neighbor's septic system.

Characteristic	Minimum	Maximum	Median	Mean
Year Home Built (n = 12)	1970	2014	1987	1989
Well Age (Years) (n = 10)	3	60	31	32
Well Depth (Feet) (n = 8)	150	500	250	283
Septic System Age (Years) (n = 10)	6	60	31	32

 Table 2. Home age and plumbing and septic system characteristics reported by each household

(b)

(a)

Characteristic	Percent
	Plastic: 80%
Premise Water Pipe Material (n = 10)	Copper: 10%
	Galvanized iron: 10%
	Sediment filter: 45%
Water Treatment System $(n - 11)$	Carbon filter: 9%
water Treatment System (n = 11)	Water softener: 9%
	No treatment: 45%
Current Septic Problems* (n = 10)	30%

\* Reported septic problems include the system filling up or making the yard wet and smelly after raining and the neighbors' system leaking into the yard.

## 4.2 Change in Concentrations of Lead and Copper After Connection to Apex Water

During each sampling event, a 250-mL sample (bottle 1) was collected after a minimum of six hours of stagnation, followed immediately by a 1-L sample (bottle 2). For private well samples, median lead levels (1.03  $\mu$ g/L) in bottle 1 exceeded the 1  $\mu$ g/L recommendation from the American Academy of Pediatrics (AAP) to prevent health impacts in children, and the maximum (64.0  $\mu$ g/L) was more than four times action level for lead (Table 3). 54% of bottle 1 samples and 20% of bottle 2 samples had lead concentrations greater than the 1  $\mu$ g/L. Lead concentrations exceeded the action level in three households (7% of bottle 1 samples and 2% of bottle 2 samples). The two households with the highest lead levels in bottle 1 (2-4 times the action level) also had water with a very low pH (<6) and alkalinity (20.4 mg/L as CaCO<sub>3</sub>). Two households (17% of bottle 1 samples and 12% of bottle 2 samples) exceeded the action level for bottle 2 samples).

One study of private wells in Orange County, North Carolina with water quality characteristics similar to this study observed a similar first-draw median lead level,  $0.99 \ \mu g/L$ (Mulhern and Gibson, 2020). However, lead levels were higher in another study focusing on peri-urban neighborhoods in Wake County, North Carolina (mean = 8.19  $\mu g/L$ , compared to 3.70  $\mu g/L$  in this study) (Stillo and Gibson, 2018) and in a study of Virginia private wells (median = 4  $\mu g/L$ , 80% of first-draw samples >=1  $\mu g/L$  and 19% of samples > the action level) (Pieper et al., 2015a). Lead levels and exceedances of the action level and AAP recommendation in the Virginia study were similar to those found during the Flint, Michigan crisis when water was sourced from corrosive Flint River water without corrosion control (Pieper et al., 2018b). A somewhat lower percentage (10%) of private well samples exceeded the action level for copper in other peri-urban neighborhoods in Wake County (Stillo and Gibson, 2018). The median, firstdraw copper level for private wells in Virginia was much higher (153 ug/L) than in this study (Pieper et al., 2015a).

		Priva	te Wells		Apex Water			
Species	%₀ >MRL <sup>a</sup>	Mean ± SD (μg/L) <sup>b</sup>	Median (µg/L) <sup>c</sup>	Maximum (µg/L)	<b>%</b> >MRL <sup>a</sup>	$\begin{array}{c} \text{Mean} \pm \\ \text{SD} \\ \left(\mu g/L\right)^b \end{array}$	Median (µg/L) <sup>c</sup>	Maximum (µg/L)
				Bottle 1				
Lead	95%	3.89 ± 11.0	1.03	64.0	73%	2.18 ± 2.02	0.472	6.20
Copper	100%	568 ± 1,291	35.7	7,070	100%	27.1 ± 35.1	15.2	165
Bottle 2								
Lead	93%	1.66 ± 3.89	0.702	22.3	67%	1.99 ± 2.10	0.451	6.34
Copper	100%	543 ± 1,816	30.5	11,400	100%	31.8 ± 40.9	16.1	192

**Table 3.** Summary statistics for lead and copper in bottles 1 and 2 for private well samples (n = 41) and Apex water samples (n = 30)

<sup>*a*</sup>MRL: method reporting limit (0.2  $\mu$ g/L for lead; 0.1  $\mu$ g/L for copper)

<sup>*b*</sup>Mean  $\pm$  SD includes samples >MRL.

<sup>*c*</sup>Median includes all samples.

Regression analysis results for lead and copper in bottles 1 and 2 are shown in Table 4.

Lead decreased significantly (p < 0.001) for bottle 1 (72%) and bottle 2 (70%) after connection to Apex water (see Figure 2(a)). Lead in 47% of bottle 1 samples and 27% of bottle 2 samples were greater than the AAP recommendation, similar to private well water results. No Apex water samples exceeded the action level for lead. Copper also decreased significantly (p < 0.001) for bottle 1 (90%) and bottle 2 (83%) (see Figure 2(b)), and no Apex water samples exceeded the action level for copper.

**Table 4.** Results of regression analysis for lead and copper. Coefficients represent effect of water source on log metal concentrations.

	Metal	Bottle	Variable	Coefficient	
	Lead	1 2	Water	$-1.27 (0.20)^{a}$ $-1.20 (0.14)^{a}$	-
	Copper	1 2	source	-2.34 (0.27) <sup><i>a</i></sup> -1.76 (0.28) <sup><i>a</i></sup>	-
	<sup>a</sup> Statistic	cally signif	ficant ( <i>p</i> <0.0	001)	
(a) 64 - 32 - 16 - 8 - 4 - 2 - 2 - 1 - 0.5 - 0.25 -	Private     Apex W	Well ater	1000 100 100 10 10 1	(b) 0- 0- 0- 1-	
	Bottle	Ζ		Bot	∠ ttle

**Figure 2.** Comparison between private well water and Apex water for concentrations of (a) lead and (b) copper in bottles 1 and 2 (n = 41 for private wells; n = 30 for Apex water)

Figure 3 displays lead concentrations in bottle 1 during the entire study period for households that connected to Apex water. Due to the COVID-19 pandemic, I was unable to resume sampling until June, so I did not measure lead concentrations immediately after connection. Lead levels one week after connection for households J and N and six weeks after connection for households E, Q, and P were generally either the same (for the household with nondetectable lead before connection) or substantially lower than private well levels. Levels were relatively stable during subsequent sampling events, with approximately 0.1 to 0.3  $\mu$ g/L fluctuations between most events. For households E and Q, which had first-draw lead 1 to 2 times the action level while on private wells, some larger fluctuations (1-3  $\mu$ g/L) occurred after connection to Apex water.

In contrast to this study where lead levels initially decreased and then remained stable, lead levels in another study continued to decrease over 30 weeks of orthophosphate dosing. Similar to this study, levels stayed low, even when pH decreased from greater than 10 to below 9.7 (Bae et al., 2020). After full lead service line replacements, lead levels have decreased below pre-replacement levels within two weeks to one month (Katner et al., 2018; Trueman et al., 2016) and continued to decrease throughout six months of post-replacement sampling (Trueman et al., 2016). Unlike this study, spikes in first-draw lead above pre-replacement levels have been observed during the first week after service line replacement (Katner et al, 2018) and one to three months after replacement (Trueman et al., 2016).



**Figure 3.** Lead concentrations in bottle 1 during the study period for the five households that connected to Apex water. Households E, Q, and P connected on May 20th, and households J and N connected on July 6th.

The average pH of Apex water (9.04) was substantially higher than in private wells (7.56) (Table 5). These results align with previous findings that higher pH and corrosion control with orthophosphate can substantially decrease lead and copper levels (Bae et al., 2020; Deshommes et al., 2018; Lytle et al., 1996). In other Wake County peri-urban neighborhoods, lead levels in private well samples were three times as high as nearby households on municipal water with corrosion control (Stillo and Gibson, 2018). Six months after reconnection to the Detroit water system and addition of orthophosphate, lead levels in Flint homes with copper service lines had decreased to levels similar to this study (median = 1.2 ug/L; 51% of samples > 1 ug/L) (Pieper et al., 2018b). Orthophosphates reduce lead levels by forming protective phosphate-rich layers on pipes, either alone or in combination with lead and other metals (Bae et al., 2020).

Measures of corrosion and scaling potential (Table 5) could also help explain changes in lead levels after connection to Apex water. Private well water and Apex water are both mildly corrosive based on the aggressive index (AI = 10-11.9) (Ahmed et al., 2021) and are at a higher risk of galvanic corrosion based on the chloride to sulfate mass ratio (CSMR >0.5 or CSMR >0.2 if alkalinity is <50 mg/L as CaCO<sub>3</sub>) (Masten et al., 2016; Nyugen et al., 2010), but the Larson Skold index (LSK<0.8) (Masten et al., 2016) indicates that they are not corrosive to iron and mild steel. Both sources of water are undersaturated with calcium carbonate and are likely to dissolve mineral scale instead of forming a protective scale according to the Langelier saturation index (LSI <0) (Masten et al., 2016). Overall, higher corrosivity of well water demonstrated by higher AI and CSMR values helps explain higher lead levels in private wells compared to Apex water.

The pH (average of 9.04, with three samples >10) and alkalinity (50-100 mg/L as CaCO<sub>3</sub>) for Apex water samples was high compared to typical values measured at the treatment plant or within the distribution system (typical pH = 7-8, typical alkalinity = 30-50 mg/L as CaCO<sub>3</sub>). Most high pH and alkalinity values were observed when the Town of Apex was not conducting hydrant flushing (see section 2: Description of Study Community and Municipal Water System) and free chlorine levels were lower due to depletion of disinfectant residuals in the distribution system. The pH and alkalinity of Apex water samples tended to be closer to typical treatment plant and distribution system values during continuous or intermittent flushing on July 27th and in September. More typical pH and alkalinity during flushing events could be related to higher free chlorine levels in the distribution system. A significant increase in pH at the tap compared to the building entrance has been observed previously, and in one study was attributed to mineral scale dissolving in the pipe and water heater (Ley et al., 2020; Salehi et al., 2020). However, it is unlikely that higher pH and alkalinity values in this study were caused by scale dissolution after the switch to Apex water because LSI values indicate that mineral scale probably did not accumulate on pipes while homes were on private wells.

	рН	Total Alkalinity (mg/L as CaCO <sub>3</sub> )	Hardness (mg/L as CaCO <sub>3</sub> )	Conductivity (µs/cm)	AI	CSMR	LSK	LSI
Private	7.56	87.7	20 (122)	240(+28)	11.2	2.15	0.03	-1.05
Wells	(±0.88)	(±18.0)	30 (±22)	249 (±38)	(±0.2)	$(\pm 0.40)$	(±0.01)	(±0.14)
Apex	9.04	48.5	26(11)	277 (+52)	10.7	0.48	0.55	-1.04
Water	(±0.89)	(±10.5)	50 (±1)	$211(\pm 33)$	(±0.4)	(±0.00)	(±0.06)	(±0.41)

**Table 5.** Basic water quality parameters and measures of corrosivity and scaling potential: mean (±SD)

# 4.3 Change in Fraction of Lead in Dissolved Form After Connection to Apex Water

The fraction of dissolved lead (averaged for all households) for private well samples was 79% in bottle 1 and 72% in bottle 2, meaning that the majority of lead was in the dissolved form instead of the particulate form (Figure 4). Dissolved lead in private well samples was significantly correlated (p < 0.001) with dissolved copper ( $\rho = 0.76-0.81$ ), zinc ( $\rho = 0.85-0.82$ ), and nickel ( $\rho = 0.62-0.75$ ) in bottles 1 and 2, indicating that brass from the faucet was likely the source of dissolved lead in most homes (Lytle et al., 2019; Pieper et al., 2018a). For two households, samples with the highest cadmium levels also had higher lead and zinc levels, indicating that galvanized pipes could be a source of dissolved lead for those homes (Clark et al., 2014; Pieper et al., 2018a). In contrast to this study, the fraction of dissolved lead in first-draw samples from private wells was approximately 50% in Virginia (Pieper et al., 2015a), and less than 50% in a North Carolina study (median = 5%) (Pieper et al., 2018a).



**Figure 4.** Dissolved lead versus total lead (dissolved + particulate) in bottles 1 and 2 for private wells compared to Apex water. Samples with detectable, total lead ( $\geq 0.2 \ \mu g/L$ ) are included.

After connection to Apex water, the fraction of lead in dissolved form decreased significantly (p < 0.001) for bottles 1 and 2 (Table 6) meaning that a higher fraction was in the particulate form. In contrast to private well results, the fraction of lead in the dissolved form was 31% in bottle 1 and 27% in bottle 2 for Apex water. Particulate lead in Apex water samples was significantly correlated (p < 0.05) with particulate aluminum ( $\rho = 0.40$ ), iron ( $\rho = 0.49$ ), chromium ( $\rho = 0.56$ ), nickel ( $\rho = 0.40$ ), and copper ( $\rho = 0.71$ ) in bottle 1 and with iron ( $\rho = 0.43$ ) and copper ( $\rho = 0.75$ ) in bottle 2. Particulate lead in these samples could be from various sources, including brass faucets and iron scales that accumulated multiple metals over time (Bae et al., 2020; Pieper et al., 2018b; Trueman et al., 2016). Aluminum in Apex water could also be from aluminum-based coagulants used in the treatment process (Li et al., 2020). First-draw lead results from 11 water utilities align with this study's results: the majority of lead from community water systems was in the particulate form, while lead in samples without corrosion control was mostly dissolved (McNeill and Edwards, 2004). Orthophosphate dosing can decrease dissolved lead levels almost immediately (Bae et al., 2020) and reduce both dissolved and particulate lead in samples dominated by particulate lead (McNeill and Edwards, 2004).

Total lead levels were much lower in Apex water than in private wells, so exposure is lower. However, it is worth noting that a large fraction of lead particulates dissolve in the digestive tract and are therefore bioavailable, and some particles can be retained in the digestive tract and continue to release lead (Triantafyllidou et al., 2007). In-vitro bioavailability of particles ranges from 2% for solder (particles composed 49% of lead) to 58-84% for brass (composed 2-7% of lead) (Deshommes and Prevost, 2012). Another concern is that the EPA method of preserving metals samples with 0.15% HNO<sub>3</sub> instead of a more concentrated 2% solution can substantially underestimate particulate lead levels compared to the amount of particulate lead that is bioavailable. However, for most water systems, especially those with non-corrosive water and lead below the action level, the EPA method adequately estimates lead levels (Triantafyllidou et al., 2007).

**Table 6.** Results of regression analysis for dissolved versus particulate lead. Coefficients represent effect of water source on the fraction of lead in dissolved form.

Bottle	Variable	Coefficient
1	Water	$-0.49 (0.06)^{a}$
2	source	-0.33 (0.07) <sup><i>a</i></sup>
~~ · ·		(

<sup>*a*</sup> Statistically significant (*p* <0.001)

#### 4.4 Change in Concentrations of Other Metals After Connection to Apex Water

#### 4.4.1 Metals of Health Concern

Four other metals of health concern changed significantly after connection to Apex water. Three of these metals, chromium, antimony, and arsenic, have MCLs intended to prevent health impacts. There is no MCL for vanadium, but there is a health recommendation from the Word Health Organization. The mean, median, and maximum values for vanadium and chromium in private well samples were all less than less than 1  $\mu$ g/L in bottles 1 and 2, and antimony and arsenic levels were all less than approximately 2  $\mu$ g/L (Table 7). After connection, there were significant increases (Table 8) in chromium in bottle 1 (51%, p = 0.0020) and bottle 2 (33%, p = 0.040), vanadium in bottle 1 (31%, p = 0.0095) and bottle 2 (41%, p < 0.001), and arsenic in bottle 1 (161%, p < 0.001) and bottle 2 (149%, p < 0.001). However, levels of these three metals remained low after connection, with mean, median, and maximum levels below 2 µg/L. Antimony also increased significantly in bottle 1 (142%, p < 0.001) and bottle 2 (132%, p < 0.001). Mean and median antimony values remained low after connection, but the maximum for bottle 1 (3.42 µg/L) was more than half of the 6 µg/L MCL and in bottle 2 (4.39 µg/L), nearly three quarters of the MCL.

#### 4.4.2 Metals of Aesthetic Concern

Four metals with MCLs intended to prevent aesthetic effects, zinc, manganese, aluminum, and iron, changed significantly after connection to Apex water. For private wells, the maximum zinc level in bottle 1 (6,450  $\mu$ g/L) was 30% higher than the 5,000  $\mu$ g/L MCL established to prevent metallic-tasting water, and 2% of samples exceeded the MCL. However, the median zinc levels (59.6  $\mu$ g/L in bottle 1 and 21.7  $\mu$ g/L in bottle 2) were much lower than then MCL. The maximum manganese level in bottle 1 (68  $\mu$ g/L) was 36% higher than the 50  $\mu$ g/L MCL established to prevent discolored or metallic-tasting water, and the maximum in bottle 2 (181  $\mu$ g/L) was more than 3.5 times the MCL. 2% of bottle 1 and 2 samples exceeded the MCL. The median manganese value in bottle 1 (1.16  $\mu$ g/L) and bottle 2 (1.21  $\mu$ g/L) was much lower than the MCL. The maximum aluminum levels in bottle 1 (593  $\mu$ g/L) and bottle 2 (510  $\mu$ g/L) were 2.5-3 times the 200  $\mu$ g/L MCL to prevent discolored water, and 7% of bottle 1 (10.2  $\mu$ g/L) and bottle 2 (6.39  $\mu$ g/L) were substantially below the MCL. For iron, the maximum level in bottle 1 (160  $\mu$ g/L) was 50% of the 300  $\mu$ g/L MCL to prevent rust-colored, metallic-

tasting water, and the maximum in bottle 2 (554  $\mu$ g/L) was almost twice the MCL. 2% of bottle 2 samples exceeded the MCL. Similar to aluminum results, median iron levels in bottle 1 (2.05  $\mu$ g/L) and bottle 2 (2.52  $\mu$ g/L) were substantially below the MCL.

Zinc and manganese decreased significantly (p < 0.001) in bottle 1 (90% for zinc and 86% for manganese) and bottle 2 (82% for zinc and 90% for manganese). Median zinc levels decreased to 21.7  $\mu$ g/L in bottle 1 and 6.82  $\mu$ g/L in bottle 2, a small fraction of the 5,000  $\mu$ g/L MCL. Maximum levels (120  $\mu$ g/L in bottle 1 and 284 in bottle 2) were 2-6% of the MCL. Similarly, median manganese levels (0.665  $\mu$ g/L in bottle 1 and 0.588 in bottle 2) were a small fraction of the 50  $\mu$ g/L MCL, and maximum manganese levels (4.58  $\mu$ g/L in bottle 1 and 2.45  $\mu$ g/L in bottle 2) were 5-9% of the MCL. Iron increased significantly in bottle 1 (141%, p =0.0013) and bottle 2 (84%, p = 0.031), as did aluminum in bottle 1 (146%, p = 0.023) and bottle 2 (136%, p = 0.0048). Median aluminum values (24.4 µg/L in bottle 1 and 22.3 µg/L in bottle 2) were only 11-12% of the MCL, but maximum values (881  $\mu$ g/L in bottle 1 and 186  $\mu$ g/L in bottle 2) were approximately 1-4.4 times the MCL. More exceedances of the aluminum MCL occurred in Apex water (17% in bottle 1 and 20% in bottle 2) than in private well samples. Median iron values (28.3 µg/L in bottle 1 and 28.6 µg/L in bottle 2) were higher than in private well samples, but were only 9% of the MCL. Maximum iron values (62.9 µg/L in bottle 1 and 86.9 µg/L in bottle 2) were lower than in private well samples and were approximately 21-29% of the MCL.

		Priva	ate Wells		Apex Water						
Metal	%₀ >MRL <sup>a</sup>	$Mean \\ \pm SD \\ (\mu g/L)^b$	Median (µg/L) <sup>c</sup>	Maximum (µg/L)	%₀ >MRL <sup>a</sup>	$Mean \\ \pm SD \\ (\mu g/L)^b$	Median (µg/L) <sup>c</sup>	Maximum (µg/L)			
Metals of Health Concern											
Bottle 1											
Vanadium	95%	$0.423 \pm 0.258$	0.298	0.941	100%	0.363 ± 0.209	0.343	1.22			
Chromium	27%	$0.199 \pm 0.085$	<mrl< td=""><td>0.337</td><td>90%</td><td><math>0.277 \pm 0.240</math></td><td>0.157</td><td>0.834</td></mrl<>	0.337	90%	$0.277 \pm 0.240$	0.157	0.834			
Antimony	22%	$0.296 \pm 0.347$	<mrl< td=""><td>1.07</td><td>100%</td><td><math>0.676 \pm 0.919</math></td><td>0.217</td><td>3.42</td></mrl<>	1.07	100%	$0.676 \pm 0.919$	0.217	3.42			
Arsenic	88%	$0.706 \pm 0.621$	0.276	2.01	100%	$0.480 \pm 0.297$	0.358	1.46			
		1		Bottle 2	1	1					
Vanadium	90%	$0.438 \pm 0.272$	0.297	0.918	100%	$0.342 \pm 0.137$	0.339	0.730			
Chromium	37%	0.221 ± 0.131	<mrl< td=""><td>0.486</td><td>93%</td><td><math>0.296 \pm 0.266</math></td><td>0.172</td><td>0.929</td></mrl<>	0.486	93%	$0.296 \pm 0.266$	0.172	0.929			
Antimony	20%	$0.368 \pm 0.626$	<mrl< td=""><td>1.90</td><td>100%</td><td><math>0.654 \pm 0.990</math></td><td>0.203</td><td>4.39</td></mrl<>	1.90	100%	$0.654 \pm 0.990$	0.203	4.39			
Arsenic	85%	$0.745 \pm 0.656$	0.255	2.10	100%	$0.454 \pm 0.289$	0.344	1.55			
			Metals	of Aesthetic C	Concern						
	<b>-</b>	1		Bottle 1	r	T					
Zinc	100%	$360 \pm 1,163$	59.6	6,450	100%	32.7 ± 31.9	21.7	120			
Iron	100%	16.4 ± 31.2	2.05	160	100%	27.2 ± 12.9	28.3	62.6			
Aluminum	100%	69.6 ± 129	10.2	593	100%	131 ± 236	24.4	881			
Manganese	100%	3.83 ± 10.8	1.16	68.0	100%	$0.901 \pm 0.870$	0.665	4.58			
		1		Bottle 2	1	1					
Zinc	100%	67.7 ± 148	24.2	871	100%	24.0 ± 52.9	6.82	284			
Iron	100%	30.7 ± 88.3	2.52	554	100%	29.9 ± 14.4	28.6	86.9			
Aluminum	100%	35.7 ± 101	6.39	510	100%	35.4 ± 40.9	22.3	186			
Manganese	95%	7.06 ± 28.9	1.21	181	100%	0.741 ± 0.479	0.588	2.45			

**Table 7.** Summary statistics for other metals in bottles 1 and 2 for private well samples (n = 41) and Apex water samples (n = 30)

<sup>*a*</sup>MRL: method reporting limit (0.5  $\mu$ g/L for aluminum; 0.1  $\mu$ g/L for all other metals) <sup>*b*</sup>Mean  $\pm$  SD includes samples >MRL. <sup>*c*</sup>Median includes all samples.

Metal	Bottle	Variable	Coefficient								
Metals of Health Concern											
Vanadium	1		$0.27 (0.10)^{a}$								
v allaululli	2		$0.34 (0.09)^{b}$								
Chromium	1		$0.41 (0.13)^{a}$								
Cinomium	2	Water	0.29 (0.14) <sup><i>c</i></sup>								
Antimony	1	source	$0.88(0.11)^{b}$								
Antimony	2		$0.84 (0.09)^{b}$								
Amonio	1		0.96 (0.12) <sup>b</sup>								
Arsenic	2		0.91 (0.12) <sup>b</sup>								
Ν	Aetals of	Aesthetic Co	oncern								
Zina	1		-2.32 (0.42) <sup>b</sup>								
Zinc	2		-1.69 (0.37) <sup>b</sup>								
Inon	1		$0.88 (0.26)^a$								
поп	2	Water	0.61 (0.28) <sup>c</sup>								
Aluminum	1	source	0.90 (0.39) <sup>c</sup>								
Alullillulli	2		0.86 (0.30) <sup><i>a</i></sup>								
Mongonoso	1		$-1.99(0.28)^{b}$								
wranganese	2		-2.26 (0.31) <sup>b</sup>								

**Table 8.** Results of regression analysis for other metals. Coefficients represent effect of water source on log metal concentrations.

<sup>a</sup>Statistically significant (p < 0.01) <sup>b</sup>Statistically significant (p < 0.001) <sup>c</sup>Statistically significant (p < 0.05)

# 4.5 Effectiveness of Flushing to Reduce Exposure to Lead

## 4.5.1 Significant Decrease in Lead Levels After-15 Second Flush

Sequential sampling results for lead are displayed in Figure 5 for homes that remained on private wells during the study (a) and homes that connected to Apex water (b). Lead levels decreased significantly (Table 9) after approximately 15 seconds (1.25 L) of flushing both for private wells (p < 0.001) and Apex water (p = 0.012). Lead levels, aggregated for all sampling events and households, decreased by 46% for private wells and 26% for Apex water. Sequential sampling results for private wells in North Carolina and Virginia also demonstrated that faucets contribute significantly to lead exposure in tap water and that lead levels decreased significantly

after the first-draw sample (Pieper et al., 2018a; Mulhern and Gibson, 2020). A risk of elevated lead levels after 15 seconds of flushing, however, was still present for some homes with private wells. Lead levels in one home still exceeded the action level, and lead in four homes (one third of all participants) was still greater than 1  $\mu$ g/L for at least one profiling event. The risk was lower for Apex water samples: no homes exceeded the action level after 15 seconds of flushing, and two homes still had lead above 1  $\mu$ g/L for at least one profiling event.

**Table 9.** Results of regression analysis for change in lead after 15 seconds (1.25 L) of flushing.

 Coefficients represent effect of bottle number on log lead concentrations.

Water Source	Variable	Coefficient					
Private wells	D - 441 - #	$-0.62 (0.13)^a$					
Apex water	Bottle #	$-0.30(0.11)^{a}$					
<sup><i>a</i></sup> Statistically significant ( $p < 0.001$ )							

Statistically significant (p (01001)

4.5.2 Flush Time and Volume to Decrease Below EPA Action Level and AAP Recommendation

For the two homes (E and P) with first-draw lead levels approximately 1 to 2 times the 15  $\mu$ g/L action level while on private wells, it took approximately 15 seconds (1.25 L) to decrease below the action level. Similar results were found for other wells in North Carolina with similar first-draw lead levels (Mulhern and Gibson, 2020). One home (D) with first-draw lead more than four times the action level took much longer, 7 minutes (33 L), to decrease below 15  $\mu$ g/L. In contrast, in another study of North Carolina wells, lead for most homes with first-draw levels several times the EPA action level decreased below 15  $\mu$ g/L within two liters of flushing (Pieper et al., 2018a). No Apex water samples had lead levels higher than the action level, including homes with first-draw levels exceeding the action level before connection. One home (P), which exceeded the action level when relying a private well, replaced the kitchen faucet before the first Apex water sampling event. The faucet replacement could have further reduced lead levels for first-draw Apex water samples compared to private well samples in this home.

One household (E) with first-draw lead levels slightly above the action level took approximately 7 minutes (33 L) to decrease below the 1  $\mu$ g/L AAP recommendation when using a private well. Homes with first-draw lead below the action level on private well water took substantially less time, 15-45 seconds (1.25-3.25 L). Only homes with first-draw levels above the action level while on private wells had first-draw levels greater than 1  $\mu$ g/L after connection to Apex water. One of these households, E, took approximately 2 minutes (9 L) to decrease below 1  $\mu$ g/L. The household that replaced its faucet, P, took 30 seconds (2.25 L).

#### 4.5.3 Sustained Lead Levels Throughout Flushing

While connected to private wells, lead levels in four homes (one third of all participating households) initially decreased during the first 15 seconds to two minutes (1.25-9 L) of flushing, followed by sustained, detectable levels below the 1  $\mu$ g/L AAP recommendation. Most (3/4) of these homes had first-draw lead below the action level. The two homes with the highest first-draw levels (2-4 times the EPA action level) had sustained lead above 1  $\mu$ g/L. Sustained lead in these homes was 77-95% dissolved and also contained elevated zinc and copper, indicating that the source could be brass components dissolving in stagnant well water (Pieper et al., 2015b). Lead was also sustained above 1  $\mu$ g/L after extensive flushing for private wells with first-draw levels above the action level in other studies in North Carolina (Mulhern and Gibson, 2020) and Virginia (Pieper et al., 2015b).

Two homes with first-draw lead levels above the action level while on private wells had sustained lead levels throughout flushing after connection to Apex water. One home, E, had sustained lead levels above  $1 \mu g/L$  for one event and between the MRL (0.2  $\mu g/L$ ) and  $1 \mu g/L$  for the second event. The other home (P), which replaced the kitchen faucet, decreased below the MRL after approximately 7 minutes (33 L) of flushing.

# 4.5.4 Spikes in Lead Levels

In private wells samples, only one spike in lead (approximately 1  $\mu$ g/L) was observed, for one household (E). The spike occurred after approximately 30 seconds (2.25 L) of flushing, likely within the premise plumbing. Lead in this sample was 89% particulate and particulate iron also increased, indicating detachment of scale as a possible source (Bae et al., 2016). Larger spikes (1-70  $\mu$ g/L) have been observed in other studies of private wells in North Carolina and Virginia, either in the particulate form as in this study (Pieper et al., 2018a; Pieper et al., 2015b) or in the dissolved form (Mulhern and Gibson, 2020).

In Apex water, one substantial spike (5.5  $\mu$ g/L) occurred for one household (Q) after approximately 2 minutes (9 L) of flushing, likely within the service line or water main. Lead in this sample was 98% particulate, and a large increase in particulate iron along with increases in particulate aluminum, chromium, manganese, nickel, copper, and zinc were observed in this sample. Particulate lead in this sample was likely from random/semi-random detachment of iron scale (Masters et al., 2016) that accumulated multiple metals over time (Trueman et al., 2016), and particulate aluminum could also be from coagulants used in the treatment process. Several homes in Flint also experienced sporadic spikes in particulate lead after reconnection to Detroit water, but in contrast to this study, spikes were several times the action level. Risk of lead spikes was likely higher in Flint because lead service lines were present in the city (Pieper et al., 2018b). Overall, the risk of increased lead exposure due to spikes in lead during flushing was low, both for private wells and Apex water.





(b) Households that connected to Apex water







**Figure 5**. Lead profiles resulting from sequential sampling in (a) households that remained on private wells during the study and (b) households that connected to Apex water. Approximate flush volume is based on the average flow rate (4.6 L/min) for faucets without aerators installed, for homes connected to private wells and Apex water.

#### **5. RESULTS FOR PFAS**

The average, total PFAS concentration (sum of all species) for samples with detectable PFAs was more than twice as high in Apex water (23.8 ng/L) compared to private wells (11.3 ng/L) (Table 10). However, this difference was not significant (coefficient = 4.24 (2.68), p =0.13). A seasonal trend in Apex water results was observed, with higher total PFAS during midsummer (mean  $\pm$  SD = 31.8  $\pm$  1.6 ng/L) compared to early fall (mean  $\pm$  SD = 15.7  $\pm$  10.9 ng/L). A recent study similarly found that the total PFAS concentration in City of Cary tap water, which is also distributed from the Cary/Apex treatment plant, was several times higher than the concentration in private wells in central North Carolina (Herkert et al., 2020). The median total PFAS concentration (8 ng/L) for private wells in that study was substantially higher than in the Irongate neighborhood (0.52 ng/L), but the maximum, total PFAS concentration was twice as high in Irongate (30.5 ng/L). Municipal water concentrations were also higher in the City of Cary study (1.5 times as high) and in 2020 monitoring results conducted by the Cary/Apex treatment plant (twice as high), compared to Apex water samples. Similar to this study, a seasonal trend in PFAS concentrations was observed in raw intake water at the Cary/Apex treatment plant during 2020. This trend is likely caused by dilution of contaminants in Jordan Lake from heavier rain during fall compared to summer.

Five PFAS species were detected in private wells, while seven were detected in Apex water.<sup>1</sup> Four of these species were perfluoroalkyl carboxylic acids (PFCAs), and four were

<sup>&</sup>lt;sup>1</sup> PFAS species detected in private wells, Apex water, or both include four perfluoroalkyl carboxylic acids (perfluorooctanoic acid (PFOA), perfluorohexanoic acid (PFHxA), perfluoropentanoic acid (PFPeA), and perfluorobutanoic acid (PFBA)) and four perfluoroalkane sulfonic acids (perfluorooctane sulfonic acid (PFOS),

perfluoroalkane sulfonic acids (PFSAs). In contrast, samples downstream of a PFAS manufacturing facility in the Cape Fear River in eastern North Carolina were dominated by GenX and other novel perfluoroether carboxylic acids (PFECAs) and perfluoroether sulfonic acids (PFESAs) (Strynar et al., 2015; Sun et al., 2016). The three most frequently detected species in private wells were PFHxSK, PFBS, and PFPeS (Table 10) and in Apex water were PFBA, PFBS, and PFPeA. Species detected in at least one private well but not in Apex water were PFPeS, PFOA, and PFOS. PFBA was detected in all Apex water samples, but in no private well samples. For two households that shared the same well, PFOS and PFPeA were each only detected in one household, and all other species were the same for the two households. Five wells had no detectable PFAS. Compared to households with total PFAS >1 ng/L, a lower percentage of households with total PFAS <1 ng/L reported septic problems (14%, compared to 40%), but average septic age was similar, approximately 30 years. Private wells were deeper (average of 320 feet) and somewhat newer (average of 28 years) for households with total PFAS <1 ng/L compared to households with total PFAS >1 ng/L (average of 260 feet and 35 years). None of these differences in household characteristics were significant (two-sample t-test, p >0.05).

potassium perfluorohexane sulfonate potassium (PFHxSK), perfluoropentane sulfonic acid (PFPeS), and perfluorobutanesulfonic acid (PFBS)).

		Pri	vate Wells		Apex Water					
Species	% >MRL <sup>a</sup>	$Mean  \pm SD  (ng/L)b$	Median (ng/L) <sup>c</sup>	Maximum (ng/L)	<b>%</b> >MRL <sup>a</sup>	$Mean  \pm SD  (ng/L)b$	Median (ng/L) <sup>c</sup>	Maximum (ng/L)		
PFOA	17%	$7.59 \pm 0.05$	<mrl< td=""><td>7.62</td><td>0%</td><td><mrl< td=""><td><mrl< td=""><td><mrl< td=""></mrl<></td></mrl<></td></mrl<></td></mrl<>	7.62	0%	<mrl< td=""><td><mrl< td=""><td><mrl< td=""></mrl<></td></mrl<></td></mrl<>	<mrl< td=""><td><mrl< td=""></mrl<></td></mrl<>	<mrl< td=""></mrl<>		
PFOS	8%	7.04 ± 0.0	<mrl< td=""><td>7.04</td><td>0%</td><td><mrl< td=""><td><mrl< td=""><td><mrl< td=""></mrl<></td></mrl<></td></mrl<></td></mrl<>	7.04	0%	<mrl< td=""><td><mrl< td=""><td><mrl< td=""></mrl<></td></mrl<></td></mrl<>	<mrl< td=""><td><mrl< td=""></mrl<></td></mrl<>	<mrl< td=""></mrl<>		
PFHxA	17%	8.12 ± 1.41	<mrl< td=""><td>9.12</td><td>60%</td><td>8.24 ± 1.08</td><td>7.25</td><td>9.97</td></mrl<>	9.12	60%	8.24 ± 1.08	7.25	9.97		
PFHxSK	58%	3.15 ± 2.61	0.52	8.15	50%	1.07 ± 0.06	0.49	1.13		
PFPeA	8%	$\begin{array}{c} 7.34 \pm \\ 0.0 \end{array}$	<mrl< td=""><td>7.34</td><td>70%</td><td>9.72 ± 0.68</td><td>9.51</td><td>10.7</td></mrl<>	7.34	70%	9.72 ± 0.68	9.51	10.7		
PFPeS	33%	0.96 ± 0.24	<mrl< td=""><td>1.20</td><td>0%</td><td><mrl< td=""><td><mrl< td=""><td><mrl< td=""></mrl<></td></mrl<></td></mrl<></td></mrl<>	1.20	0%	<mrl< td=""><td><mrl< td=""><td><mrl< td=""></mrl<></td></mrl<></td></mrl<>	<mrl< td=""><td><mrl< td=""></mrl<></td></mrl<>	<mrl< td=""></mrl<>		
PFBA	0%	<mrl< td=""><td><mrl< td=""><td><mrl< td=""><td>100%</td><td>8.51 ± 1.60</td><td>8.28</td><td>12.6</td></mrl<></td></mrl<></td></mrl<>	<mrl< td=""><td><mrl< td=""><td>100%</td><td>8.51 ± 1.60</td><td>8.28</td><td>12.6</td></mrl<></td></mrl<>	<mrl< td=""><td>100%</td><td>8.51 ± 1.60</td><td>8.28</td><td>12.6</td></mrl<>	100%	8.51 ± 1.60	8.28	12.6		
PFBS	42%	1.42 ± 0.31	<mrl< td=""><td>1.67</td><td>100%</td><td>2.96 ± 1.60</td><td>3.41</td><td>4.56</td></mrl<>	1.67	100%	2.96 ± 1.60	3.41	4.56		
Total PFAS	100%	$11.3 \pm 12.3$	0.52	30.5	58%	23.8 ± 11.3	30.7	34.3		

**Table 10.** Summary statistics for PFAS in private well samples (n = 12) and Apex water samples (n = 10)

<sup>*a*</sup>MRL: method reporting limit (6.24 ng/L for PFOA, PFOS, PFPeA, and PFHxA; 0.5 ng/L for PFHxSK, PFPeS, PFBA, and PFBS

<sup>*b*</sup>Mean  $\pm$  SD includes samples >MRL.

<sup>*c*</sup>Median includes all samples.

The most frequently detected species were not always the largest contributors to PFAS sample composition. Private well samples were composed mostly of PFHsK, followed by PFHxA and PFOA (Figure 6). Apex water samples were composed mostly of PFBA, PFPeA, and PFHxA. In previous analyses of tap water from the Cary/Apex Water Treatment Plant, contributions from PFHxA, PFHxSK, PFPeA were similar to results from Irongate, but PFBA and PFBS contributions were somewhat lower (Herkert et al., 2020; Monschein, 2021). Different PFAS concentrations and species composition reported previously for water from the Cary/Apex treatment plant could be partially attributed to the lower method detection limit (MDL) used to report results from those analyses compared to the MRL used in this study. For example, PFOA, PFOS, and PFHpA were detected by the treatment plant in 2020 at

concentrations below this study's MRL (6.24 ng/L) for those species. The presence of PFOA and PFOS in private wells and samples from the Cary/Apex treatment plant mirror previous results in North Carolina and elsewhere in the United States (Guelfo et al., 2018; Sun et al., 2016) demonstrating that these compounds remain in water, despite a voluntary agreement by manufacturers to eliminate PFOA in emissions and products by 2015 (U.S. EPA, 2021) and to phase out PFOS by 2002 (Buck et al., 2011).

Three long-chain PFAS (PFOA, PFOS and PFHxSK) defined as having eight or more carbons for PFCAs and six or more for PFSAs (ITRC, 2020), were detected in the Irongate neighborhood. Private well water composition was half PFCAs and half PFSAs, and the percentage of long-chain species (56%) was somewhat higher than short-chain species. Apex water was dominated by PFCAs (85%) and short-chain species (98%). A higher fraction of PFCAs in surface water compared to groundwater was previously found and attributed to surface sources of PFCAs including manufacturing, landfills, and wastewater treatment plants (Guelfo et al., 2018).

Removal in water systems and behavior in the human body are different for the PFCAs and short-chain species that dominate Apex water compared to PFSAs and long-chain species. PFSAs and long-chain PFAS are easier to remove with activated carbon home treatment systems (Herkert et al., 2020) than PFCAs and short-chain species. There is evidence that PFSAs and long-chain PFAS are less likely to bioaccumulate (Conder et al., 2008) and that long-chain PFAS remain in the body longer as demonstrated by their longer serum half-life (Xu et al., 2020). However, short-chain PFAS could have toxicity similar to long-chain PFAS (Gomis et al., 2018) and are just as persistent in the environment (Brendel et al., 2018).



**Figure 6.** PFAS composition (% of total PFAS) for private wells (n = 12 households; n = 12 samples) and Apex water (n = 5 households; n = 10 samples)

Table 11 lists health advisories and MCLs in effect as of March 2021 for PFAS species detected in Irongate. State health advisories and MCLs are included if they differ from EPA's health advisory level (70 ng/L for the sum of PFOA and PFOS). Health advisories are nonenforceable levels above which actions such as taking a water source out of use or notifying the public may be taken. In contrast, MCLs are enforceable standards that public water systems must comply with. The most commonly researched species, PFOA, PFOS, and PFHxS, are also the species most commonly included in health advisories and MCLs. PFBS is addressed by three states, and PFHxA and PFBA by only one state. PFPeA and PFPeS are not addressed by any health advisories or MCLs. PFAS addressed by zero to three states make up 44% of the species composition of private wells in the Irongate neighborhood and 98% of Apex water. No exceedances of health advisory levels or MCLs were observed for private wells or Apex water. However, concentrations in the well that served two households were 76-95% of PFOA advisory levels or MCLs in three states, 70% of New York's MCL for PFOS, 92% of Massachusetts' MCL for the sum of six species, and 92% of Vermont's MCL for the sum of five species. In contrast, all Apex water samples were 0% of state MCLs and advisory levels for PFOA and PFOS, and half of samples were approximately 5% of Massachusetts and Vermont's MCLs.

A non-exhaustive list of use categories for PFAS detected in the Irongate neighborhood is also shown in Table 11. Most species have not been used intentionally in products or processes but were detected analytically, likely as impurities or degradants (Glüge et al., 2020). The highest number of species have been detected in AFFF (8 species), wood construction materials and leather (7 species), and floor polish and food packaging (6 species). Species detected in Irongate have been found in diverse use categories. Uses for PFAS species with the highest number of detections in private well samples and that contribute most to sample composition include AFFF, apparel, leather, wood construction products, photographic materials, metal products, food packaging, bakeware, floor polish, workwear, plastic and rubber production, and semi-conductor processes. Additional use categories for PFOA include cosmetics, dental floss, electronic devices, pesticides, paints, ski wax, textiles, and auto parts. Use categories for species commonly detected or abundant in Apex water include AFFF, food packaging, cosmetics, electronic devices, apparel, leather, floor polish, wood construction products, ski wax, textiles, photographic materials, workwear, plastic/rubber production, semi-conductor, dental floss, and metal products.

Research on the health effects of PFAS is evolving, and most studies have focused on the "legacy" species PFOA and PFOS, along with PFHxS. Health effects listed in Table 11 for the species found in Irongate are not exhaustive and are included based on the weight of evidence across studies. PFOA and PFOS, which were detected in one well, are associated with a wide range of health effects including thyroid disorders (Lopez-Espinoza et al., 2012; Blake et al., 2018), disrupted weight regulation (Liu et al., 2018), higher cholesterol (Frisbee et al., 2010;

Nelson et al., 2010), lower immune response to routine vaccines (Grandjean et al., 2012; Granum et al., 2013; Looker et al., 2014; Stein et al., 2016), and pregnancy-induced hypertension (Darrow et al., 2013). PFOA is additionally associated with kidney and testicular cancer (Barry et al., 2013; Vieira et al., 2013) and ulcerative colitis (Steenland et al. 2013). PFHxS, which was detected in 60% of private well samples and 50% of Apex water samples, is associated with decreased kidney function (Blake et al., 2018), liver damage, disrupted weight regulation (Liu et al., 2018), and reduced immune response to vaccines (Grandjean et al., 2012; Granum et al., 2013; Stein et al., 2016). PFBA, which is found in all Apex water samples and makes up the highest percentage (36%) of species composition, is associated with higher severity of COVID-19 (Grandjean et al., 2020). For the remaining species detected in Irongate, evidence is either inconclusive or cannot be extrapolated to humans.

C	PFOS	PFOA	PFHxSK	PFHxA	PFPeS	PFPeA	PFBS	PFBA
Chain Length	8		6		5		4	
Sources								
AFFF	U	D	U	D	D	D	D	D
Food packaging	D	D		D		D	D	D
Bakeware		D					D	
Cosmetics		D		D		D		D
Dental Floss		D				D		
Electronic devices	U	U				U		D
Apparel	D	D	D	D		D	D	D
Leather	D	D	D	D		D	D	D
Floor polish	D	D		D		D	D	D
Pesticides		D						
Paints		U						
Wood construction materials	D	D	D	D	D	D		D
Ski wax		D		D		D		D
Textiles		D		D		D		D

**Table 11.** Carbon chain length, sources, health effects, and health advisory levels or enforceable MCLs for the eight PFAS species detected in the Irongate neighborhood

	PFOS	PFOA	PFHxSK	PFHxA	PFPeS	PFPeA	PFBS	PFBA
Sources				1		1		1
Photographic								••
materials	U	U	D					U
Workwear								
for medical		D		D			D	
staff, pilots		D		D			D	
of firemen								
Plastic and								
rubber							U	
production								
Chrome	TT							
plating	U							
Semi-						TT	TT	
conductor						U	U	
Metal			T.					
products			U					
Auto parts		U						
Aviation								
fluids and	U							
fuels	Ũ							
Health Effects								
Kidney and	, 							
testicular		1						
cancer		•						
Decreased								
kidney			<ul> <li>✓</li> </ul>					
function								
Disrupted								
thyroid	~	$\checkmark$						
function	-	•						
Illcerative								
colitis		$\checkmark$						
Liver damage	✓	<ul> <li>✓</li> </ul>	×					
Interformed	•	•	-					
with woight	1	1	1					
regulation	•	•	·					
High								
cholesterol	✓	$\checkmark$						
Programancy								
induced	1	$\checkmark$						
hypertension	•	•						
Reduced								
immune								
response to	✓	$\checkmark$	✓					
vaccines								
Increased								
severity of								✓
COVID-19								

	PFOS	PFOA	PFHxSK	PFHxA	PFPeS	PFPeA	PFBS	PFBA
Health Adviso	ory (ng/L)							
EPA	70 (P	FOS +						
	PF	OA)						
California	40	10						
Connecticut	70 (PFC	A + PFOS	+ PFNA +					
	PF	FHxS + PFF	HpA)					
Minnesota	15	35	47				2,000	7,000
Ohio	70 (PFOS + PFOA)		140				140.000	
							140,000	
MCL (ng/L)								
Mass.	20 (PFC	A + PFOS	+ PFNA +					
	PFHxS	+ PFHpA	+ PFDA)					
Michigan	16	8	51	400,000			420	
New	15	10	19					
Hampshire		12	10					
New Jersey	13	14						
New York	10	10						
Vermont	20 (PFOA + PFOS + PFNA +							
	PF	FHxS + PFI	HpA)					

D = detected analytically; U = used currently or historically

### 6. CONCLUSION

This is the first study to assess the change in drinking water quality after an underbounded community relying on private wells connected to municipal water. Overall, connection to municipal water in this neighborhood reduced exposure to lead and copper, which are associated with many harmful health impacts. Lead levels remained low during the study instead of spiking during subsequent sampling events as seen after lead service line replacement. In this neighborhood, running the tap for short periods of time effectively reduced lead exposure for both private wells and households connected to municipal water. However, a risk of sustained exposure, even with extensive flushing, existed in homes with first-draw lead above EPA's action level. For those homes, it may be advisable to install a filter to reduce lead levels in addition to flushing. Although not statistically significant, the average, total PFAS concentration was higher in municipal water than in private wells. The composition of PFAS species after connection shifted to a higher percentage of PFCAs and short-chained species, which may be less bioaccumulative but are more difficult to remove from carbon filters used in homes. PFAS species in both water sources are likely to be toxic to humans. Efforts should be made to continue to reduce PFAS levels in municipal water and to develop more stringent drinking water standards, and private wells in North Carolina should continue to be monitored to provide more data on the prevalence of PFAS.

# **APPENDIX: HOUSEHOLD SURVEY**

Questions about your well, your plumbing, and your water

These questions will help us keep track of where any contaminants are coming from. They will help us figure out if contamination goes away when the Apex water connections are in place. If you do not feel comfortable answering certain questions, you may leave them blank.

# **Questions**

When was this home built (year)? \_\_\_\_\_

Do you have a water treatment or filtration system for your whole house or kitchen tap?

- 🛛 No
- Yes
  - Treats water for the whole house
  - Treats water for kitchen tap

Туре

- Treats water for the whole house
- $\circ$   $\;$  Treats water for kitchen tap
- Water softener
- $\circ \quad \text{Iron filter} \quad$
- $\circ \quad \text{Sediment filter} \\$
- Carbon filter
- UV disinfection
- Constant chlorination
- o Reverse osmosis
- o Don't know

If you have a water treatment or filtration system, please briefly describe how you maintain it (for example, changing filters or adding salt to a water softener).

About how old is your well? \_\_\_\_\_ years

About how deep is your well? \_\_\_\_\_\_ feet

Have you had any problems with your well running dry or not providing enough water during the past year? If yes, please describe any problems.

About how old is your septic system? \_\_\_\_\_\_ years

Is your septic system currently causing problems? Please describe any current problems.

Has your septic system had problems in the past? Please describe any past problems.

What material is the water pipes in your house? If you're not sure, please look at the pipes connected to your kitchen sink (see the arrow pointing to the water supply line in the picture below for an example).

- Copper (copper or orange-colored metal piping, could be shiny or dull)
- Plastic (white, cream, blue, red, or gray piping)
- Galvanized iron (gray or silver-gray metal)
- o Other
- I don't know



Please help us measure the flow rate of your kitchen faucet by following the steps below. Place a liquid measuring cup that holds at least two cups underneath your kitchen faucet. If you don't have a large measuring cup, you can use a large cup or bottle and then transfer the water to a smaller measuring cup to measure the amount. Please <u>do not use your water</u> <u>sampling bottles</u> for this task.

- 1. Turn the faucet on all the way and at the same time, start a timer on a stopwatch or phone.
- 2. Turn the faucet off after 5 seconds.
- 3. Record the amount of water (ounces, cups, or milliliters) that you collected.

\_\_ounces <u>OR</u> \_\_\_\_\_cups <u>OR</u> \_\_\_\_\_milliliters

When we sampled your water last March, was an aerator installed on your kitchen faucet? An aerator is a small device you can screw on the end of a faucet, or an aerator might already be installed on your faucet when you buy it. You might see a small screen on the end of the faucet if an aerator is installed. They reduce the amount of water coming out and make it splash less.

- 🛛 No
- Yes

Did you make any changes to your water filtration or treatment system after the last sampling event you participated in? If yes, please describe.

Have you performed any maintenance on your water filtration or treatment system since the last sampling event you participated in? If yes, please describe.

Have you made any changes to the plumbing <u>inside your home</u> or to your faucet since the last sampling event you participated in? If yes, please describe.

Has your home been hooked up to Apex water service?

🛛 No

Yes

If yes, on what date was your home hooked up?

If no, do you still plan to connect your water to Apex water service?

- 🛛 No
- Yes

Thank you for taking the time to answer these questions!

#### REFERENCES

21 Vt. Code R. 12 030 003-6.12 (2021)

310 Mass. Code Regs. 22.07G(3)

- Ahmed, S., Sultan, M. W., Alam, M., Hussain, A., Qureshi, F., & Khurshid, S. (2021). Evaluation of corrosive behaviour and scaling potential of shallow water aquifer using corrosion indices and geospatial approaches in regions of the Yamuna river basin. *Journal of King Saud University - Science*, 33(1), 101237. https://doi.org/10.1016/j.jksus.2020.101237
- Aiken, C. S. (1987). Race as a factor in municipal underbounding. *Annals of the Association of American Geographers*, 77(4), 564–579. https://doi.org/10.1111/j.1467-8306.1987.tb00181.x
- Apex, NC. Ordinance 2020-0121-07. (Jan. 21, 2020).
- Bae, Y., Pasteris, J. D., & Giammar, D. E. (2020). Impact of orthophosphate on lead release from pipe scale in high pH, low alkalinity water. *Water Research*, 177, 115764. https://doi.org/10.1016/j.watres.2020.115764
- Barry, V., Winquist, A., & Steenland, K. (2013). Perfluorooctanoic acid (PFOA) exposures and incident cancers among adults living near a chemical plant. *Environmental Health Perspectives*, 121(11–12), 1313–1318. https://doi.org/10.1289/ehp.1306615
- Barton, K. E., Starling, A. P., Higgins, C. P., McDonough, C. A., Calafat, A. M., & Adgate, J. L. (2020). Sociodemographic and behavioral determinants of serum concentrations of perand polyfluoroalkyl substances in a community highly exposed to aqueous film-forming foam contaminants in drinking water. *International Journal of Hygiene and Environmental Health*, 223(1), 256–266. https://doi.org/10.1016/j.ijheh.2019.07.012
- Bates, D., Mächler, M., Bolker, B., & Walker, S. (2015). Fitting linear mixed-effects models using lme4. *Journal of Statistical Software*, 67(1), 1–48. https://doiorg.libproxy.lib.unc.edu/10.18637/jss.v067.i01
- Bentel, M. J., Yu, Y., Xu, L., Li, Z., Wong, B. M., Men, Y., & Liu, J. (2019). Defluorination of per- and polyfluoroalkyl substances (PFASs) with hydrated electrons: Structural dependence and implications to PFAS remediation and management. *Environmental Science & Technology*, 53(7), 3718–3728. https://doi.org/10.1021/acs.est.8b06648
- Blake, B. E., Pinney, S. M., Hines, E. P., Fenton, S. E., & Ferguson, K. K. (2018). Associations between longitudinal serum perfluoroalkyl substance (PFAS) levels and measures of thyroid hormone, kidney function, and body mass index in the Fernald Community Cohort. *Environmental Pollution*, 242(Pt A), 894–904. https://doi.org/10.1016/j.envpol.2018.07.042

- Brendel, S., Fetter, É., Staude, C., Vierke, L., & Biegel-Engler, A. (2018). Short-chain perfluoroalkyl acids: Environmental concerns and a regulatory strategy under REACH. *Environmental Sciences Europe*, 30(1), 9. https://doi.org/10.1186/s12302-018-0134-4
- Buck, R. C., Franklin, J., Berger, U., Conder, J. M., Cousins, I. T., de Voogt, P., Jensen, A. A., Kannan, K., Mabury, S. A., & van Leeuwen, S. P. J. (2011). Perfluoroalkyl and polyfluoroalkyl substances in the environment: Terminology, classification, and origins. *Integrated Environmental Assessment and Management*, 7(4), 513–541. https://doi.org/10.1002/ieam.258
- California Water Boards. CA Water Boards. (2020, February 12). *Notification level issuance: PFOA*. <u>https://www.waterboards.ca.gov/drinking\_water/certlic/drinkingwater/documents/pfos\_a</u> <u>nd\_pfoa/pfoa\_nl\_issuance\_jan2020.pdf</u>
- California Water Boards. CA Water Boards. (2020, February 14). *Notification level issuance: PFOS.* <u>https://www.waterboards.ca.gov/drinking\_water/certlic/drinkingwater/documents/pfos\_a</u> <u>nd\_pfoa/pfos\_nl\_issuance\_jan2020.pdf</u>
- Centers for Disease Control and Prevention (CDC). (2015, July 1). *Lead and drinking water from private wells*. Retrieved February 22, 2021, from <u>https://www.cdc.gov/healthywater/drinking/private/wells/disease/lead.html</u>
- Clark, B.N., Masters, S.V., and Edwards, M.A. (2014). Lead release to drinking water from galvanized steel pipe coatings. *Environmental Engineering Science* 32:8, 713-721. https://doi.org/10.1089/ees.2015.0073
- Conder, J. M., Hoke, R. A., De Wolf, W., Russell, M. H., & Buck, R. C. (2008). Are PFCAs bioaccumulative? A critical review and comparison with regulatory criteria and persistent lipophilic compounds. *Environmental Science & Technology*, 42(4), 995–1003. https://doi.org/10.1021/es070895g

Control of Lead and Copper, 40 C.F.R. §141.80-141.91 (1991).

- Darrow, L. A., Stein, C. R., & Steenland, K. (2013). Serum perfluorooctanoic acid and perfluorooctane sulfonate concentrations in relation to birth outcomes in the Mid-Ohio Valley, 2005-2010. *Environmental Health Perspectives*, 121(10), 1207–1213. https://doi.org/10.1289/ehp.1206372
- Delgado, C. F., Ullery, M. A., Jordan, M., Duclos, C., Rajagopalan, S., & Scott, K. (2018). Lead exposure and developmental disabilities in preschool-aged children. *Journal of public health management and practice: JPHMP*, 24(2), e10–e17. https://doi.org/10.1097/PHH.00000000000556

- Deshommes, E., Trueman, B., Douglas, I., Huggins, D., Laroche, L., Swertfeger, J., Spielmacher, A., Gagnon, G. A., & Prévost, M. (2018). Lead levels at the tap and consumer exposure from legacy and recent lead service line replacements in six utilities. *Environmental Science & Technology*, 52(16), 9451–9459. https://doi.org/10.1021/acs.est.8b02388
- Deshommes, E., Laroche, L., Deveau, D., Nour, S., & Prévost, M. (2017). Short- and long-term lead release after partial lead service line replacements in a metropolitan water distribution system. *Environmental Science & Technology*, 51(17), 9507–9515. https://doi.org/10.1021/acs.est.7b01720
- Deshommes, E., & Prévost, M. (2012). Pb particles from tap water: Bioaccessibility and contribution to child exposure. *Environmental Science & Technology*, 46(11), 6269– 6277. https://doi.org/10.1021/es2045585
- Dietrich, A. M., Glindemann, D., Pizarro, F., Gidi, V., Olivares, M., Araya, M., Camper, A., Duncan, S., Dwyer, S., Whelton, A.J., Younos, T., Subramanian, S., Burlingame, G.A., Khiari, D., & Edwards, M. (2004). Health and aesthetic impacts of copper corrosion on drinking water. *Water Science and Technology*, 49(2), 55-62. Retrieved from http://libproxy.lib.unc.edu/login?url=https://www-proquestcom.libproxy.lib.unc.edu/scholarly-journals/health-aesthetic-impacts-copper-corrosionon/docview/1943310888/se-2?accountid=14244
- Dvorak, B.I., & Skipton, S.O. (2008). *EC08-703 Drinking water treatment: An overview*. Historical materials from University of Nebraska-Lincoln Extension. http://digitalcommons.unl.edu/extensionhist/4869
- Fox, M. A., Nachman, K. E., Anderson, B., Lam, J., & Resnick, B. (2016). Meeting the public health challenge of protecting private wells: Proceedings and recommendations from an expert panel workshop. *Science of the Total Environment*, 554–555, 113–118. https://doi.org/10.1016/j.scitotenv.2016.02.128
- Fram, M. S., & Belitz, K. (2011). Occurrence and concentrations of pharmaceutical compounds in groundwater used for public drinking-water supply in California. *The Science of the Total Environment*, 409(18), 3409–3417. https://doi.org/10.1016/j.scitotenv.2011.05.053
- Frisbee, S. J., Shankar, A., Knox, S. S., Steenland, K., Savitz, D. A., Fletcher, T., & Ducatman, A. M. (2010). Perfluorooctanoic acid, perfluorooctanesulfonate, and serum lipids in children and adolescents: results from the C8 Health Project. *Archives of Pediatrics & Adolescent Medicine*, 164(9), 860–869. https://doi.org/10.1001/archpediatrics.2010.163
- Gibson, J. M., Fisher, M., Clonch, A., MacDonald, J. M., & Cook, P. J. (2020). Children drinking private well water have higher blood lead than those with city water. *Proceedings of the National Academy of Sciences of the United States of America*, 117(29), 16898–16907. https://doi.org/10.1073/pnas.2002729117

- Gibson, J.M., DeFelice, N., Sebastian, D., & Leker, H. (2014). Racial disparities in access to community water supply service in Wake County, North Carolina. *American Journal of Public Health*, 104(12), e45–e45. https://doi.org/10.2105/AJPH.2014.10412e45.1
- Ginsberg, G., & Toal, B. (2016, December 12). Drinking water action level for perfluorinated alkyl substances (PFAS). Connecticut Department of Health. <u>https://www.asdwa.org/wp-content/uploads/2018/01/CT-PFASActionLevel.pdf</u>
- Glüge, J., Scheringer, M., Cousins, I. T., DeWitt, J. C., Goldenman, G., Herzke, D., Lohmann, R., Ng, C. A., Trier, X., & Wang, Z. (2020). An overview of the uses of per- and polyfluoroalkyl substances (PFAS). *Environmental Science. Processes & Impacts*, 22(12), 2345–2373. https://doi.org/10.1039/d0em00291g
- Gomis, M. I., Vestergren, R., Borg, D., & Cousins, I. T. (2018). Comparing the toxic potency in vivo of long-chain perfluoroalkyl acids and fluorinated alternatives. *Environment International*, 113, 1–9. https://doi.org/10.1016/j.envint.2018.01.011
- Gomis, M. I., Wang, Z., Scheringer, M., & Cousins, I. T. (2015). A modeling assessment of the physicochemical properties and environmental fate of emerging and novel per- and polyfluoroalkyl substances. *Science of the Total Environment*, 505, 981–991. https://doi.org/10.1016/j.scitotenv.2014.10.062
- Goodlad, J. K., Marcus, D. K., & Fulton, J. J. (2013). Lead and attention-deficit/hyperactivity disorder (ADHD) symptoms: a meta-analysis. *Clinical Psychology Review*, *33*(3), 417–425. https://doi.org/10.1016/j.cpr.2013.01.009
- Grandjean, P., Timmermann, C. A. G., Kruse, M., Nielsen, F., Vinholt, P. J., Boding, L., Heilmann, C., & Mølbak, K. (2020). Severity of COVID-19 at elevated exposure to perfluorinated alkylates. *Plos One*, 15(12), e0244815. https://doi.org/10.1371/journal.pone.0244815
- Grandjean, P., Andersen, E. W., Budtz-Jørgensen, E., Nielsen, F., Mølbak, K., Weihe, P., & Heilmann, C. (2012). Serum vaccine antibody concentrations in children exposed to perfluorinated compounds. *The Journal of the American Medical Association*, 307(4), 391–397. https://doi.org/10.1001/jama.2011.2034
- Granum, B., Haug, L. S., Namork, E., Stølevik, S. B., Thomsen, C., Aaberge, I. S., van Loveren, H., Løvik, M., & Nygaard, U. C. (2013). Pre-natal exposure to perfluoroalkyl substances may be associated with altered vaccine antibody levels and immune-related health outcomes in early childhood. *Journal of Immunotoxicology*, 10(4), 373–379. https://doi.org/10.3109/1547691X.2012.755580
- Guelfo, J. L., & Adamson, D. T. (2018). Evaluation of a national data set for insights into sources, composition, and concentrations of per- and polyfluoroalkyl substances (PFASs) in U.S. drinking water. *Environmental Pollution*, 236, 505–513. https://doi.org/10.1016/j.envpol.2018.01.066

- Harari, F., Sallsten, G., Christensson, A., Petkovic, M., Hedblad, B., Forsgard, N., Melander, O., Nilsson, P. M., Borné, Y., Engström, G., & Barregard, L. (2018). Blood lead levels and decreased kidney function in a population-based cohort. *American Journal of Kidney Diseases*, 72(3), 381–389. https://doi.org/10.1053/j.ajkd.2018.02.358
- Heaney, C. D., Wing, S., Wilson, S. M., Campbell, R. L., Caldwell, D., Hopkins, B., O'Shea, S., & Yeatts, K. (2013). Public infrastructure disparities and the microbiological and chemical safety of drinking and surface water supplies in a community bordering a landfill. *Journal of Environmental Health*, 75(10), 24-36. Retrieved from http://libproxy.lib.unc.edu/login?url=https://www-proquest-com.libproxy.lib.unc.edu/scholarly-journals/public-infrastructure-disparities-microbiological/docview/1354972336/se-2?accountid=14244
- Heaney, C., Wilson, S., Wilson, O., Cooper, J., Bumpass, N., & Snipes, M. (2011). Use of community-owned and -managed research to assess the vulnerability of water and sewer services in marginalized and underserved environmental justice communities. *Journal of Environmental Health*, 74(1), 8-17. Retrieved from http://libproxy.lib.unc.edu/login?url=https://www-proquestcom.libproxy.lib.unc.edu/scholarly-journals/use-community-owned-managed-researchassess/docview/878224975/se-2?accountid=14244
- Henningsen, A. (2017). censReg: Censored Regression (Tobit) Models. R package version 0.5. http://CRAN.R-Project.org/package-censReg.
- Herkert, N. J., Merrill, J., Peters, C., Bollinger, D., Zhang, S., Hoffman, K., Ferguson, P. L., Knappe, D. R. U., & Stapleton, H. M. (2020). Assessing the effectiveness of point-of-use residential drinking water filters for perfluoroalkyl substances (PFASs). *Environmental Science & Technology Letters*, 7(3), 178-184. https://doi.org/10.1021/acs.estlett.0c00004
- Hopkins, Z. R., Sun, M., DeWitt, J. C., & Knappe, D. R. U. (2018). Recently detected drinking water contaminants: GenX and other per- and polyfluoroalkyl ether acids. *Journal -American Water Works Association*, 110(7), 13–28. https://doi.org/10.1002/awwa.1073
- Hu, X. C., Andrews, D. Q., Lindstrom, A. B., Bruton, T. A., Schaider, L. A., Grandjean, P., Lohmann, R., Carignan, C. C., Blum, A., Balan, S. A., Higgins, C. P., & Sunderland, E. M. (2016). Detection of poly- and perfluoroalkyl substances (PFASs) in U.S. drinking water linked to industrial sites, military fire training areas, and wastewater treatment plants. *Environmental Science & Technology Letters*, *3*(10), 344–350. https://doi.org/10.1021/acs.estlett.6b0026
- Interstate Technology and Regulatory Council (ITRC). (2020, April). Naming conventions and physical and chemical properties of per- and polyfluoroalkyl substances (PFAS). Retrieved March 1, 2021, from <u>https://pfas-</u> 1.itrcweb.org/fact\_sheets\_page/PFAS\_Fact\_Sheet\_Naming\_Conventions\_April2020.pdf

- Johnson, J. H., Parnell, A., Joyner, A. M., Christman, C. J., & Marsh, B. (2004). Racial apartheid in a small North Carolina town. *The Review of Black Political Economy*, *31*(4), 89–107. https://doi.org/10.1007/s12114-004-1012-4
- Katner, A., Pieper, K., Brown, K., Lin, H.-Y., Parks, J., Wang, X., Hu, C.-Y., Masters, S., Mielke, H., & Edwards, M. (2018). Effectiveness of prevailing flush guidelines to prevent exposure to lead in tap water. *International Journal of Environmental Research* and Public Health, 15(7). https://doi.org/10.3390/ijerph15071537
- Kotlarz, N., Collier, D., Lea, S., Knappe, D., & Hoppin, J. (2019). Population exposure to emerging per- and polyfluoroalkyl substances (PFAS) through drinking water in North Carolina. *Environmental Epidemiology*, *3*, 214. doi: 10.1097/01.EE9.0000608200.05204.5c
- Kwiatkowski, C. F., Andrews, D. Q., Birnbaum, L. S., Bruton, T. A., DeWitt, J. C., Knappe, D. R. U., Maffini, M. V., Miller, M. F., Pelch, K. E., Reade, A., Soehl, A., Trier, X., Venier, M., Wagner, C. C., Wang, Z., & Blum, A. (2020). Scientific basis for managing PFAS as a chemical class. *Environmental Science & Technology Letters*. https://doi.org/10.1021/acs.estlett.0c00255
- Lanphear, B. P., Rauch, S., Auinger, P., Allen, R. W., & Hornung, R. W. (2018). Low-level lead exposure and mortality in US adults: A population-based cohort study. *The Lancet. Public Health*, *3*(4), e177–e184. https://doi.org/10.1016/S2468-2667(18)30025-2
- Ley, C. J., Proctor, C. R., Jordan, K., Ra, K., Noh, Y., Odimayomi, T., Julien, R., Kropp, I., Mitchell, J., Nejadhashemi, A. P., Whelton, A. J., & Aw, T. G. (2020). Impacts of municipal water-rainwater source transitions on microbial and chemical water quality dynamics at the tap. *Environmental Science & Technology*, 54(18), 11453–11463. https://doi.org/10.1021/acs.est.0c03641
- Li, G., Bae, Y., Mishrra, A., Shi, B., & Giammar, D. E. (2020). Effect of aluminum on lead release to drinking water from scales of corrosion products. *Environmental Science & Technology*, 54(10), 6142–6151. https://doi.org/10.1021/acs.est.0c00738
- Lichter, D. T., Parisi, D., Grice, S. M., & Taquino, M. (2007). Municipal underbounding: Annexation and racial exclusion in small southern towns. *Rural Sociology*, 72(1), 47–68. https://doi.org/10.1526/003601107781147437
- Lifetime Health Advisories and Health Effects Support Documents for Perfluorooctanoic Acid and Perfluorooctane Sulfonate, 81 Fed Reg. 33,250 (May 25, 2016).
- Lin, Y., Huang, L., Xu, J., Specht, A. J., Yan, C., Geng, H., Shen, X., Nie, L. H., & Hu, H. (2019). Blood lead, bone lead and child attention-deficit-hyperactivity-disorder-like behavior. *Science of the Total Environment*, 659, 161–167. https://doi.org/10.1016/j.scitotenv.2018.12.219

- Liu, G., Dhana, K., Furtado, J. D., Rood, J., Zong, G., Liang, L., Qi, L., Bray, G. A., DeJonge, L., Coull, B., Grandjean, P., & Sun, Q. (2018). Perfluoroalkyl substances and changes in body weight and resting metabolic rate in response to weight-loss diets: A prospective study. *PLoS Medicine*, 15(2), e1002502. https://doi.org/10.1371/journal.pmed.1002502
- Lockhart, S., Wood, E., & MacDonald Gibson, J. (2020). Impacts of exclusion from municipal water service on water availability: A case study. *New Solutions*, *30*(2), 127–137. https://doi.org/10.1177/1048291120932913
- Looker, C., Luster, M. I., Calafat, A. M., Johnson, V. J., Burleson, G. R., Burleson, F. G., & Fletcher, T. (2014). Influenza vaccine response in adults exposed to perfluorooctanoate and perfluorooctanesulfonate. *Toxicological Sciences*, 138(1), 76–88. https://doi.org/10.1093/toxsci/kft269
- Lopez-Espinosa, M.-J., Mondal, D., Armstrong, B., Bloom, M. S., & Fletcher, T. (2012). Thyroid function and perfluoroalkyl acids in children living near a chemical plant. *Environmental Health Perspectives*, 120(7), 1036–1041. https://doi.org/10.1289/ehp.1104370
- Lytle, D. A., Schock, M. R., Wait, K., Cahalan, K., Bosscher, V., Porter, A., & Del Toral, M. (2019). Sequential drinking water sampling as a tool for evaluating lead in Flint, Michigan. *Water Research*, 157, 40–54. https://doi.org/10.1016/j.watres.2019.03.042
- Lytle, D., Schock, M., & Sorg, T.J. (1996). Controlling lead corrosion in the drinking water of a building by orthophosphate and silicate treatment. *Jour. New England Water Works Assoc*, 110(3), 202-210.
- Marsh, B., Parnell, A. M., & Joyner, A. M. (2010). Institutionalization of racial inequality in local political geographies. *Urban Geography*, *31*(5), 691–709. https://doi.org/10.2747/0272-3638.31.5.691
- Masten, S. J., Davies, S. H., & Mcelmurry, S. P. (2016). Flint water crisis: What happened and why? *Journal American Water Works Association*, *108*(12), 22–34. https://doi.org/10.5942/jawwa.2016.108.0195
- McNeill, L. S., & Edwards, M. (2004). Importance of Pb and Cu particulate species for corrosion control. *Journal of Environmental Engineering*, *130*(2), 136–144

Mich. Admin. Code r.325.10604(g)

Minnesota Department of Health. (n.d.). *Perfluoroalkyl substances (PFAS)*. Retrieved March 1, 2021, from <a href="https://www.health.state.mn.us/communities/environment/hazardous/topics/pfcs.html#safelevels">https://www.health.state.mn.us/communities/environment/hazardous/topics/pfcs.html#safelevels</a>

- Miranda, M. L., Kim, D., Galeano, M. A., Paul, C. J., Hull, A. P., & Morgan, S. P. (2007). The relationship between early childhood blood lead levels and performance on end-of-grade tests. *Environmental Health Perspectives*, 115(8), 1242–1247. https://doi.org/10.1289/ehp.9994
- Monschein, R. (2021, February 10). *Cary/Apex water treatment facility fact sheet: Per and polyfluoroalkyl substances (PFAS)*. Town of Cary. Retrieved March 1, 2021, from <a href="https://www.townofcary.org/home/showpublisheddocument?id=23001">https://www.townofcary.org/home/showpublisheddocument?id=23001</a>
- Monschein, R. (2020, April 8). *Cary/Apex water treatment facility fact sheet: Per and polyfluoroalkyl substances (PFAS)*. Town of Cary. Retrieved December 8, 2020, from <u>https://www.townofcary.org/home/showdocument?id=23001</u>
- Mulhern, R., & Gibson, J.M. (2020). Under-sink activated carbon water filters effectively remove lead from private well water for over six months. *Water*, *12*(12), 3584. https://doi.org/10.3390/w12123584

N.C. Gen. Stat. § 160D-202(a)

- Nelson, J. W., Hatch, E. E., & Webster, T. F. (2010). Exposure to polyfluoroalkyl chemicals and cholesterol, body weight, and insulin resistance in the general U.S. population. *Environmental health perspectives*, 118(2), 197–202. https://doi.org/10.1289/ehp.0901165
- N.H. Rev. Stat. Ann. § 485:16(e) (2020)
- N.J. Admin. Code § 7:10-5.2(a)(5)(ii)–(iii) (2020)
- North Carolina Department of Environmental Quality (NC DEQ) (2019). *Local water supply planning*. Retrieved December 8, 2020, from <u>https://www.ncwater.org/WUDC/app/LWSP/report.php?pwsid=03-92-045&year=2019</u>
- Nguyen, C. K., Stone, K. R., & Edwards, M. A. (2011). Chloride-to-sulfate mass ratio: Practical studies in galvanic corrosion of lead solder. *Journal / American Water Works Association*, 103(1), 81-92. doi:10.1002/j.1551-8833.2011.tb11384.x
- N.Y. Comp. Codes R. & Regs. tit. 10, § 5-1.52 (2020)
- Ohio Environmental Protection Agency and Ohio Department of Health. (n.d.). *PFAS technical information and supporting documentation*. <u>https://epa.ohio.gov/Portals/28/documents/pfas/PFAS-Technical-Information-Supporting-Documentation.pdf</u>
- Parnell AM, Joyner AM, Christman CJ, Marsh DP. (2004). The persistence of political segregation: Racial underbounding in North Carolina. Retrieved from http://www.cedar groveinst.org/files/regional\_underbounding.pdf.

- Pieper, K. J., Nystrom, V. E., Parks, J., Jennings, K., Faircloth, H., Morgan, J. B., Bruckner, J., & Edwards, M. A. (2018a). Elevated lead in water of private wells poses health risks: Case study in Macon County, North Carolina. *Environmental Science & Technology*, 52(7), 4350–4357. https://doi.org/10.1021/acs.est.7b05812
- Pieper, K. J., Martin, R., Tang, M., Walters, L., Parks, J., Roy, S., Devine, C., & Edwards, M. A. (2018b). Evaluating water lead levels during the Flint water crisis. *Environmental Science & Technology*, 52(15), 8124–8132. https://doi.org/10.1021/acs.est.8b00791
- Pieper, K. J., Krometis, L.-A. H., Gallagher, D. L., Benham, B. L., & Edwards, M. (2015a). Incidence of waterborne lead in private drinking water systems in Virginia. *Journal of Water and Health*, 13(3), 897–908. https://doi.org/10.2166/wh.2015.27
- Pieper, K. J., Krometis, L.-A., Gallagher, D., Benham, B., & Edwards, M. (2015b). Profiling private water systems to identify patterns of waterborne lead exposure. *Environmental Science & Technology*, 49(21), 12697–12704. https://doi.org/10.1021/acs.est.5b03174
- Purchase, J. M., Rouillier, R., Pieper, K. J., & Edwards, M. (2020). Understanding failure modes of NSF/ANSI 53 lead-certified point-of-use pitcher and faucet filters. *Environmental Science & Technology Letters*. https://doi.org/10.1021/acs.estlett.0c00709
- Reduction of Lead in Drinking Water Act, Pub. L. No. 111-380, sec. 2, 42 U.S.C. § 300g-6(d), 124 Stat. 4131 (2011).
- Richards, R. P., Baker, D. B., Creamer, N. L., Kramer, J. W., Ewing, D. E., Merryfield, B. J., & Wallrabenstein, L. K. (1996). Well water quality, well vulnerability, and agricultural contamination in the midwestern United States. *Journal of Environmental Quality*, 25(3), 389–402. https://doi.org/10.2134/jeq1996.00472425002500030002x
- Safe Drinking Water Act Amendments of 1986, Pub. L. 99-359, sec. 109, 42 U.S.C. § 300g-6(d), 100 Stat. 642 (1986).
- Saleeby, B., Shimizu, M. S., Sanchez Garcia, R. I., Avery, G. B., Kieber, R. J., Mead, R. N., & Skrabal, S. A. (2021). Isomers of emerging per- and polyfluoroalkyl substances in water and sediment from the Cape Fear River, North Carolina, USA. *Chemosphere*, 262, 128359. https://doi.org/10.1016/j.chemosphere.2020.128359
- Salehi, M., Odimayomi, T., Ra, K., Ley, C., Julien, R., Nejadhashemi, A. P., Hernandez-Suarez, J. S., Mitchell, J., Shah, A. D., & Whelton, A. (2020). An investigation of spatial and temporal drinking water quality variation in green residential plumbing. *Building and Environment*, 169, 106566. https://doi.org/10.1016/j.buildenv.2019.106566
- Schaider, L. A., Balan, S. A., Blum, A., Andrews, D. Q., Strynar, M. J., Dickinson, M. E., Lunderberg, D. M., Lang, J. R., & Peaslee, G. F. (2017). Fluorinated compounds in U.S. fast food packaging. *Environmental Science & Technology Letters*, 4(3), 105–111. https://doi.org/10.1021/acs.estlett.6b00435

- Schaider, L. A., Ackerman, J. M., & Rudel, R. A. (2016). Septic systems as sources of organic wastewater compounds in domestic drinking water wells in a shallow sand and gravel aquifer. *The Science of the Total Environment*, 547, 470–481. https://doi.org/10.1016/j.scitotenv.2015.12.081
- Schock, M. R. (1989). Understanding corrosion control strategies for lead. Journal American Water Works Association, 81(7), 88–100. https://doi.org/10.1002/j.1551-8833.1989.tb03244.x
- Shoemaker, J. A., Grimmett, P., & Boutin, B. (2008). Determination of Selected Perfluorinated Alkyl Acids in Drinking Water by Solid Phase Extraction and Liquid Chromatography/Tandem Mass Spectrometry (LC/MS/MS). U.S. Environmental Protection Agency, Washington, DC, USA.
- Sorensen, L. C., Fox, A. M., Jung, H., & Martin, E. G. (2018). Lead exposure and academic achievement: Evidence from childhood lead poisoning prevention efforts. *Journal of Population Economics*, *32*(1), 1–40. https://doi.org/10.1007/s00148-018-0707-y
- Steenland, K., Zhao, L., Winquist, A., & Parks, C. (2013). Ulcerative colitis and perfluorooctanoic acid (PFOA) in a highly exposed population of community residents and workers in the mid-Ohio valley. *Environmental Health Perspectives*, 121(8), 900– 905. https://doi.org/10.1289/ehp.1206449
- Stein, C. R., McGovern, K. J., Pajak, A. M., Maglione, P. J., & Wolff, M. S. (2016). Perfluoroalkyl and polyfluoroalkyl substances and indicators of immune function in children aged 12-19 y: National Health and Nutrition Examination Survey. *Pediatric Research*, 79(2), 348–357. https://doi.org/10.1038/pr.2015.213
- Stillo, F., & Gibson, J. M. (2018). Racial disparities in access to municipal water supplies in the American south: Impacts on children's health. *International Public Health Journal*, 10(3), 309-323. Retrieved from http://libproxy.lib.unc.edu/login?url=https://www-proquestcom.libproxy.lib.unc.edu/scholarly-journals/racial-disparities-access-municipalwater/docview/2189949033/se-2?accountid=14244
- Stillo, F., & Gibson, J.M. (2017). Exposure to contaminated drinking water and health disparities in North Carolina. *American Journal of Public Health*, 107(1), 180–185. https://doi.org/10.2105/AJPH.2016.303482
- Strynar, M., Dagnino, S., McMahen, R., Liang, S., Lindstrom, A., Andersen, E., McMillan, L., Thurman, M., Ferrer, I., & Ball, C. (2015). Identification of novel perfluoroalkyl ether carboxylic acids (PFECAs) and sulfonic acids (PFESAs) in natural waters using accurate mass time-of-flight mass spectrometry (TOFMS). *Environmental Science & Technology*, 49(19), 11622–11630. https://doi.org/10.1021/acs.est.5b01215

- Sun, M., Arevalo, E., Strynar, M., Lindstrom, A., Richardson, M., Kearns, B., Pickett, A., Smith, C., & Knappe, D. R. U. (2016). Legacy and emerging perfluoroalkyl substances are important drinking water contaminants in the Cape Fear River watershed of North Carolina. *Environmental Science & Technology Letters*, 3(12), 415–419. https://doi.org/10.1021/acs.estlett.6b00398
- Sunderland, E. M., Hu, X. C., Dassuncao, C., Tokranov, A. K., Wagner, C. C., & Allen, J. G. (2019). A review of the pathways of human exposure to poly- and perfluoroalkyl substances (PFASs) and present understanding of health effects. *Journal of Exposure Science & Environmental Epidemiology*, 29(2), 131–147. https://doi.org/10.1038/s41370-018-0094-1
- Tobin J. Estimation of relationships for limited dependent variables. *Econometrica* 1958, 24–36.
- Town of Apex. (2019). Water Quality Annual Report. <u>https://www.apexnc.org/DocumentCenter/View/1593/Annual-Drinking-Water-Quality-Report?bidId</u>)
- Town of Apex. (n.d.). *Apex utility map*. Retrieved December 8, 2020, from <u>https://apexnc.maps.arcgis.com/apps/webappviewer/index.html?id=7f61e4e9219f49019d</u> <u>01b40ff0c06e07</u>
- Town of Cary. 2019. Water Quality Testing Summary. <u>https://www.townofcary.org/home/showdocument?id=22499</u>
- Town of Cary. (n.d.). Annual water disinfection change. Retrieved December 8, 2020, from <u>https://www.townofcary.org/services-publications/water-sewer/water/water-treatment/annual-water-disinfecting-change</u>
- Triantafyllidou, S., Schock, M. R., DeSantis, M. K., & White, C. (2015). Low contribution of PbO2-coated lead service lines to water lead contamination at the tap. *Environmental Science & Technology*, 49(6), 3746–3754. https://doi.org/10.1021/es505886h
- Triantafyllidou, S., Parks, J., & Edwards, M. (2007). Lead particles in potable water. *American Water Works Association Journal*, *99*(6), 107-117,12.
- Trueman, B. F., Camara, E., & Gagnon, G. A. (2016). Evaluating the effects of full and partial lead service line replacement on lead levels in drinking water. *Environmental Science & Technology*, 50(14), 7389–7396. https://doi.org/10.1021/acs.est.6b01912
- U.S. Census Bureau. (2019 [race]; 2015-2019 [income]). *QuickFacts: Apex Town, North Carolina*. Retrieved November 27, 2020, from <u>https://www.census.gov/quickfacts/fact/table/apextownnorthcarolina/EDU685218#EDU6</u> <u>85217</u>

- United States Environmental Protection Agency (U.S. EPA). (2021, March 4). Assessing and managing chemicals under TSCA Fact sheet: 2010/2015 PFOA Stewardship Program. Retrieved March 4, 2021, from https://www.epa.gov/assessing-and-managing-chemicals-under-tsca/fact-sheet-20102015-pfoa-stewardship-program#what
- U.S. EPA. *PFAS Master List of PFAS Substances*. (2020). Retrieved March 18, 2021, from https://comptox.epa.gov/dashboard/chemical\_lists/pfasmaster
- U.S. EPA. (1996). Safe drinking water act amendments of 1996. Retrieved March 17, 2021, from https://www.epa.gov/sites/production/files/2020-05/documents/safe\_drinking\_water\_act-title\_xiv\_of\_public\_health\_service\_act.pdf
- U.S. EPA. (1994). Method 200.8: Determination of Trace Elements in Waters and Wastes by Inductively Coupled Plasma-Mass Spectrometry, Revision 5.4. Cincinnati, OH, USA.
- Vieira, V. M., Hoffman, K., Shin, H.-M., Weinberg, J. M., Webster, T. F., & Fletcher, T. (2013). Perfluorooctanoic acid exposure and cancer outcomes in a contaminated community: a geographic analysis. *Environmental Health Perspectives*, 121(3), 318–323. https://doi.org/10.1289/ehp.1205829
- Wilson, S. M., Heaney, C. D., Cooper, J., & Wilson, O. (2008). Built environment issues in unserved and underserved African-American neighborhoods in North Carolina. *Environmental Justice*, 1(2), 63–72. https://doi.org/10.1089/env.2008.0509
- Xu, Y., Fletcher, T., Pineda, D., Lindh, C. H., Nilsson, C., Glynn, A., Vogs, C., Norström, K., Lilja, K., Jakobsson, K., & Li, Y. (2020). Serum half-lives for short- and long-chain perfluoroalkyl acids after ceasing exposure from drinking water contaminated by firefighting foam. *Environmental Health Perspectives*, 128(7), 77004. https://doi.org/10.1289/EHP6785
- Yeung, L. W. Y., Dassuncao, C., Mabury, S., Sunderland, E. M., Zhang, X., & Lohmann, R. (2017). Vertical profiles, sources, and transport of PFASs in the Arctic Ocean. *Environmental Science & Technology*, 51(12), 6735–6744. https://doi.org/10.1021/acs.est.7b00788
- Zheng, Y., & Flanagan, S. V. (2017). The case for universal screening of private well water quality in the U.S. and testing requirements to achieve it: Evidence from arsenic. *Environmental Health Perspectives*, 125(8), 085002. https://doi.org/10.1289/EHP629