Addressing gaps in the US EPA Lead and Copper Rule: Developing guidance and improving citizen science tools to mitigate corrosion in public water systems and premise plumbing

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(ABSTRACT)

Lead and copper in drinking water are known to pose aesthetic and health concerns for humans and pets. The United States Environmental Protection Agency (US EPA) Lead and Copper Rule (LCR) set 90th percentile action levels for lead (15 ppb) and copper (1.3 mg/L), above which utilities must implement systemwide corrosion control. However, gaps in the US EPA LCR leave at least 10% of residents using municipal water and all private well users vulnerable to elevated lead and copper in their drinking water. To help address these gaps in the LCR, this dissertation 1) Evaluates accuracy of at-home lead in water test kits to help residents identify lead problems, 2) Refines orthophosphate corrosion control guidance to help reduce cuprosolvency, 3) Identifies challenges to mitigating cuprosolvency by raising pH, and 4) Develops guidance that can help residents assess and address cuprosolvency problems.

Lead in drinking water can pose a variety of health concerns, particularly for young children. The revised LCR will still leave many residents unprotected from elevated lead in their drinking water and potentially wondering what to do about it. Many consumers concerned about lead may choose to purchase at-home lead in water test kits, but there is no certification authority to ensure their accuracy. Most off-the-shelf tests purchased in this work (12 of 16) were not able to detect dissolved or particulate lead at levels of concern in drinking

water (i.e. near the lead action level of 15 ppb) due to high detection limits (5,000-20,000 ppb). Binary type tests, which indicate the presence or absence of lead based on a trigger threshold of 15 ppb, were often effective at detecting dissolved lead, but they failed to detect the presence of leaded particles that often cause high lead exposures in drinking water problems. Some of these problems detecting particles could be reduced using simple at-home acid dissolution with weak household acids such a vinegar or lemon juice. Our analysis points out the strengths and weaknesses of various types of at-home lead in water tests, which could be particularly important considering potential distrust in official results in the aftermath of the Flint Water Crisis.

Elevated cuprosolvency, or copper release into drinking water, can be an aesthetic concern due to fixture staining, blue water, and green hair and can pose health concerns for residents and pets. In addition to the general gaps in the LCR described above, compliance sampling in the LCR focuses on older homes at highest risk of elevated lead, rather than the newer homes at highest risk of elevated copper. Problems with elevated copper can sometimes go undetected as a result. Guidance was developed to help proactive utilities address cuprosolvency issues through the addition of orthophosphate corrosion inhibitors or pH adjustment as a function of a water's alkalinity. Linear regressions developed from pipe cuprosolvency tests (R^2>0.98) determined a "minimum" orthophosphate dose or a "minimum" pH for a given alkalinity that was expected to almost always reduce copper below the 1.3 mg/L EPA action level in a reasonable length of time. The subjective nature of the terms "almost always" and "reasonable length of time" were quantitatively discussed based on laboratory and field data.

Orthophosphate addition was generally very effective at cuprosolvency control. Orthophosphate treatment in copper tube cuprosolvency tests produced cuprosolvency below the action level within the first week of treatment. As expected, orthophosphate treated waters some-

times resulted in higher long-term cuprosolvency than the same waters without orthophosphate corrosion control treatment. This is consistent with the formation of phosphate scales which have an intermediate solubility between the cupric hydroxide in new pipes and the malachite or tenorite scales expected in pipe aging without orthophosphate. A linear regression (R^2>0.98) was used to determine the orthophosphate dose needed for a given alkalinity to yield copper below the 1.3 mg/L action level in the pipe segments with the highest, 2nd highest, 3rd highest copper concentrations (100th, 95th, or 90th percentile, n=20 replicates, five each from four manufacturers) after 4 or 22 weeks of pipe aging. This regression was generally in good agreement with a bin approach put forth in the 2015 Consensus Statement from the National Drinking Water Advisory Council, but in some cases the regression predicted that higher orthophosphate doses would be needed.

In contrast, due to the greater complexity of the reactions involved, a similar simplistic approach for pH adjustment is not widely applicable. A linear regression predicted that higher "minimum" pH values would be needed to control cuprosolvency compared to those suggested by the 2015 National Drinking Water Advisory Consensus Statement. Results indicate that factors such as the potential for calcite precipitation, pipe age, and significant variability in cuprosolvency from pipes of different manufacturers may warrant further research. Field LCR monitoring data indicated that 90th percentile copper concentrations continued to decline over a period of years or decades when orthophosphate is not used, and our laboratory results demonstrate a few cases where copper levels even increased with time. Consideration of confounding effects from other water quality parameters such as natural organic matter, silica, and sulfate would be necessary before the "minimum" pH criteria could be broadly applied.

Guidance was then developed to help address cuprosolvency issues on a single building or single home basis for residents with private wells or those with high copper in municipal systems meeting the LCR. A hierarchy of costs and considerations for various interventions are discussed including replumbing with alternative materials, using bottled water or point use pitcher, tap, or reverse osmosis filters to reduce copper consumption, and using whole house interventions like more conventional orthophosphate addition and pH adjustment, or unproven strategies like granular activated carbon filtration, reverse osmosis treatment, and ion exchange treatment. Laboratory and citizen science testing demonstrated that some inexpensive at-home tests for pH and copper, were accurate enough to serve as inputs for this guidance and could empower consumers to diagnose their problems and consider possible solutions. Citizen science field testing and companion laboratory studies of potential interventions indicate that short-term (<36 weeks) use of pH adjustment, granular activated carbon, anion exchange and reverse osmosis treated water were not effective at forming a protective scale for the resident's water tested. In this case-study, cuprosolvency problems were ultimately related to water chemistry and linked to variability in influent water pH.

Overall, this work highlighted weaknesses in the current US EPA Lead and Copper Rule. It attempted to close some of these gaps by assessing the accuracy of at-home citizen science tests for lead and copper detection and developing guidance to support voluntary interventions by utilities or consumers. Ideally, local authorities (utilities, health departments, cooperative extension programs) could adapt this guidance to account for local water quality considerations and support consumers in resolving cuprosolvency issues. This guidance may also serve as a citizen science approach that some consumers could use to make decisions on their own. Future work could extend and improve on these initial efforts.

Addressing gaps in the US EPA Lead and Copper Rule: Developing guidance and improving citizen science tools to mitigate corrosion in public water systems and premise plumbing

Rebecca B. Kriss

(GENERAL AUDIENCE ABSTRACT)

Lead or copper in drinking water can come from corrosion of plumbing materials. Elevated levels of these metals can cause aesthetic concerns like blue water and fixture staining, as well as health concerns for humans and pets. The United States Environmental Protection Agency (US EPA) Lead and Copper Rule (LCR) is designed to address system wide lead and copper corrosion problems in municipal water supplies. According to the LCR, utilities must notify consumers and implement corrosion control if more than 10% of homes sampled have lead above 15 ppb or copper above 1.3 mg/L. However, gaps in the US EPA LCR leave at least 10% of residents using municipal water and all private well users vulnerable to elevated lead and copper in their drinking water. To help address these gaps in the LCR, this dissertation 1) Evaluates how accurate residential at-home tests are at detecting lead in water, 2) Refines orthophosphate corrosion control guidance to help address elevated cuprosolvency (i.e. copper release to water), 3) Identifies challenges addressing cuprosolvency issues by raising the pH, and 4) Develops guidance to help residents detect and address cuprosolvency problems.

Lead in drinking water can come from corrosion of lead bearing plumbing such as lead service lines and lead solder. Lead can pose a variety of health concerns, particularly for

young children. In spite of recent revisions, the LCR will still leave many residents unprotected from elevated lead in their drinking water and potentially wondering what to do about it. Many consumers concerned about lead may choose to purchase at-home lead in water test kits, but there is no certification authority to ensure that they are accurate. Most off-the-shelf tests purchased in this work (12 of 16) were not able to detect dissolved lead or lead from particulate at concentrations expected to occur in drinking water due to high detection limits (5,000-20,000 ppb). Binary type tests, which indicate the presence or absence of lead based on a trigger threshold of 15 ppb, were often effective at detecting dissolved lead, but they failed to detect the presence of leaded particles that often cause high lead exposures in drinking water problems. Some of these problems detecting particles could be reduced using a simple procedure to attempt to dissolve the particles using weak household acids like vinegar or lemon juice. Our analysis points out the strengths and weaknesses of various types of at-home lead in water tests, which could be particularly important considering potential distrust in official results in the aftermath of the Flint Water Crisis.

Elevated cuprosolvency, or copper release into drinking water, primarily causes aesthetic problems like fixture staining and blue water, and it can also pose acute and serious health concerns for residents and some pets. Many of the same issues with the LCR that leave residents at risk of lead can also lead to unaddressed issues with elevated copper. In addition to those issues, the LCR focuses on collecting water samples in older homes at highest risk of lead, instead of newer homes at highest risk of copper. This means that many cuprosolvency problems could go undetected. Guidance was developed to help proactive utilities address cuprosolvency problems throughout the whole water system by adding orthophosphate corrosion inhibitors or adjusting the pH of their water. Linear relationships were developed from cuprosolvency testing in copper pipes (strong correlations, R^2>0.98) to determine the "minimum" orthophosphate dose or pH value needed based on the water alkalinity that was

expected to almost always reduce copper below the 1.3 mg/L EPA action level in a reasonable length of time. We also discuss the subjective nature of the terms "almost always" and "reasonable length of time" based on laboratory and field data.

Adding orthophosphate was generally very effective at controlling cuprosolvency. In tests in copper pipe segments, copper concentrations in the water were below the action level within one week of starting to add orthophosphate. As expected, sometimes waters with orthophosphate treatment resulted in higher long-term copper concentrations than waters without orthophosphate. This is in agreement with reports of formation of phosphate mineral scales which have an intermediate solubility between those in new pipes and the scales expected in pipe aging without orthophosphate. A linear regression (strong correlation, R^2>0.98) was used to determine the orthophosphate dose needed for a given alkalinity to yield copper below the 1.3 mg/L action level in the worst, second worst, and third worst pipes of the 20 pipe segments tested (100^{th}, 95^{th}, or 90^{th} percentile) after 4 or 22 weeks of pipe aging. This linear relationship was generally in good agreement with a bin approach put forth in the 2015 Consensus Statement from the National Drinking Water Advisory Council, but in some cases the regression predicted that higher orthophosphate doses would be needed.

In contrast, we showed that adjusting the pH to control cuprosolvency was too simplistic to be widely applicable because the chemical reactions involved are more complex. The linear relationship we developed predicted that higher "minimum" pH values would be needed to control cuprosolvency compared to those suggested by the 2015 National Drinking Water Advisory Consensus Statement. Other factors such as the potential calcite precipitation, which can clog pipes, pipe age, and significant variability in copper coming off pipes from different manufacturers may require consideration when considering treatment options. LCR monitoring data from utilities indicated that copper concentrations continued to decline over

a period of years or decades when orthophosphate was not used, and our laboratory results demonstrate a few cases where copper levels even increased with time. We also showed that other water quality components like natural organic matter, silica, and sulfate can affect cuprosolvency and could make it difficult to broadly apply the "minimum" pH approach for controlling cuprosolvency in places with different water qualities.

Guidance was then developed to help address cuprosolvency issues on a single building or single home basis for residents with private wells or those with high copper in municipal systems meeting the LCR. A hierarchy of costs and considerations is described for various interventions including replumbing with alternative materials, using bottled water or point use pitcher, tap, or reverse osmosis filters to reduce copper consumption, and using whole house interventions like more conventional orthophosphate addition and pH adjustment, or unproven strategies like granular activated carbon filtration, reverse osmosis treatment, and ion exchange treatment. Laboratory and citizen science testing demonstrated that some inexpensive at-home tests for pH and copper, were accurate enough to serve as inputs for this guidance and could empower consumers to diagnose their problems and consider possible solutions. Testing of potential water treatments in the laboratory and citizen science testing in a resident's home showed that short-term (<36 weeks) use of pH adjustment, granular activated carbon, anion exchange, and reverse osmosis treated water did not form a permanent, low-solubility protective scale for this resident's water. In this case-study, cuprosolvency problems were ultimately related to water chemistry and linked to variability in incoming pH of the water.

This thesis highlighted weaknesses in the current US EPA Lead and Copper Rule. It attempted to address some of these issues by determining the accuracy of at-home citizen science tests to help residents detect lead and copper and developing guidance to support

voluntary interventions by utilities or consumers. Ideally, local authorities (utilities, health departments, cooperative extension programs) could adapt this guidance to account for local water quality considerations and support consumers in resolving cuprosolvency issues. This guidance may also serve as a citizen science approach that some consumers could use to make decisions on their own. Future work could extend and improve on these initial efforts.

Dedication

This dissertation is dedicated to my family and friends.

In particular, it is dedicated to my parents, who instilled in me a value for education and caring for others, and to other loved ones we've lost. May this honor your memory.

I would also like to dedicate this to my husband, whose support and faith in me never wavered, and to those who encouraged me on this journey.

I wouldn't be who I am without your love and support.

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List of Abbreviations

AAS Atomic Absorption Spectroscopy

ACLU American Civil Liberties Union

AFM Atomic Force Microscopy

AL Action Level

ANOVA Analysis of Variance

ASV Anodic Stripping Voltammetry

CAMA 2014 Coal Ash Management Act

CCR Consumer Confidence Report

CCT Corrosion Control Treatment

Cr(VI) hexavalent chromium

ddPCR droplet digital Polymerase Chain Reaction

DEQ Department of Environmental Quality

DI Deionized

DO Dissolved Oxygen

DOC Dissolved Organic Carbon

DPB Disinfection Byproduct

FA Factor Analysis

FDA United States Food and Drug Administration

FOIA Freedom of Information Act

GAC Granuallar Activated Carbon

HREs Health Risk Evaluations

ICP-MS Inductively Coupled Plasma - Mass Spectrometry

IHSS International Humic Substance Society

LCR Lead and Copper Rule

LDH Louisiana Department of Health

LI Langelier Index

LSL Lead Service Line

MCL Maximum Contaminant Level

MDEQ Michigan Department of Environmental Quality

NCDEQ North Carolina Department of Environmental Quality

NDWAC National Drinking Water Advisory Council

NOM Natural Organic Matter

NSF/ANSI National Sanitation Foundation/American National Standards Institute

OKC Oklahoma City

OP Orthophosphate

PCA Principal Component Analysis

PLSLR Partial Lead Service Line Replacement

Poly-P Polyphosphate

POU point-of-use

ppb parts per billion

QA/QC Quality Assurance/Quality Control

qPCR quantitative Polymerase Chain Reaction

RFC Raftelis Financial Consultants

RO Reverse Osmosis

RWIC Louisiana Rural Water Infrastructure Committee

SC DHEC South Carolina Department of Health and Environmental Control

SD Standard Deviation

SDWA Safe Drinking Water Act

SMCL Secondary Maximum Contaminant Level

TDS Total Dissolved Solids

TOC Total Organic Carbon

TTHM Total Trihalomethane

US CDC United States Centers for Disease Control and Prevention

US EPA United States Environmental Protection Agency

VT Virginia Tech

WLL Water Lead Level

XRD X-Ray Diffraction

Chapter 1

Introduction

1.1 Lead and Copper in Drinking Water

1.1.1 Sources and concerns of elevated lead and copper in drinking water

Elevated lead and copper in drinking water generally result from corrosion of plumbing materials. Copper corrosion can include a variety of mechanisms such as pitting, erosion corrosion, wall thinning, and copper release to water (i.e. cuprosolvency).[284] In the context of this work, we focus on corrosion control to address cuprosolvency problems. The extent of corrosion problems will depend on a combination of factors including the presence lead bearing (lead service lines, leaded brass, lead solder) or copper plumbing, the water chemistry, the presence of corrosion inhibitors, and the age of the plumbing.[262, 304] The use of leaded plumbing (lead service line, lead solder, materials >8% lead by weight) were banned in a 1986 amendment to the Safe Drinking Water Act.[4] The amount of lead allowed in plumbing materials was further reduced in 2014 to a maximum of 0.25% by weight on the wetted surface area.[37] However, concerns remain about childhood lead exposure from legacy plumbing materials, especially in light of the recent Flint Water Crisis and the 2001-2004 Washington, DC Lead Crisis.[107, 109, 112, 117, 214] This concern sparked further efforts, via the 2021 Bipartisan Infrastructure Bill, to remove leaded plumbing materials.[31] In contrast,

relatively little attention has been paid to issues with elevated cuprosolvency which tends to occur more frequently in recently installed pipe. This is particularly concerning since copper pipes are still widely used for home plumbing and in lead service line replacements.[33]

Exposure to lead and copper in drinking water present aesthetic and health concerns. Lead, a neurotoxin linked to developmental issues, remains a significant risk to long-term human health.[142] Although other exposure pathways can also contribute to lead exposure, recent cases of water-related elevated blood lead demonstrate the importance of addressing lead in drinking water.[55, 112, 117, 142] Elevated copper in drinking water is often problematic because of aesthetic concerns like blue water and fixture staining, but can also cause acute and long-term health effects like gastrointestinal distress, liver and kidney problems, and sometimes more serious health effects for certain sensitive populations and some pets.[8, 115, 304, 329] Because of these concerns, the US EPA set unenforceable health based maximum contaminant level goals for lead (0 µg/L) and copper (1.3 mg/L) in drinking water.[36]

1.1.2 Decoupling Lead and Copper: Gaps in the Lead and Copper Rule

The US EPA regulated lead and copper concentrations in drinking water via the Lead and Copper Rule (LCR). The LCR is not designed to completely protect public health, but rather, to detect and address systemwide corrosion issues due to practices of municipal water systems. According to the LCR, utilities must address corrosion issues via public notification and corrosion control treatment (CCT) if over 10% of samples in a high risk sampling pool exceed the 90^{th} percentile action levels (AL) of 15 ppb for lead and 1.3 mg/L for copper.[8, 44, 49, 262, 339]

Gaps in the LCR could leave many residents vulnerable to elevated lead and copper in their drinking water. Because the LCR uses a 90th percentile AL, up to 10% of residents in high-risk homes could have very high lead and/or copper before public notification and corrosion control interventions are triggered. Further, the LCR primarily regulates water quality of homes in municipal systems, but private wells and other non-residential buildings are generally not regulated, even though recent updates to the LCR now requires lead sampling in schools and early childcare centers.[27, 49, 99, 304, 339] These gaps can leave all private well owners, many residents, and non-residential building owners at risk of cuprosolvency issues in their drinking water.[44, 115, 270]

Further, several weaknesses in the LCR could allow widespread corrosion problems to persist, potentially undetected, exposing even more residents to risk of elevated lead and copper. First, thousands of community systems are struggling to be in compliance with the LCR and there are cases where utilities are in compliance with the LCR and implementing CCT (phosphate inhibitors), but elevated copper persisted.[58, 239, 342] By using the high-risk lead plumbing sampling pool to monitor for both lead and copper, the LCR cannot detect the worst case cuprosolvency issues. The worst lead problems almost always occur in older homes (pre-1986) with leaded plumbing, but elevated copper is most likely in homes with newer plumbing. Therefore, the LCR is likely to underestimate or completely miss cuprosolvency issues, which will only get worse as the LCR sampling pool continues to age.[44, 303, 304, 339] The 2015 National Drinking Water Advisory Council (NDWAC) consensus statement proposed updates to the LCR to address widespread cuprosolvency issues, however, the most recent LCR revisions did not follow these reccomendations.[15, 27, 30] Even with the focus on detecting lead issues, improper sampling methods and analytical procedures that could miss particles and lead from service lines could allow water lead hazards to go undetected.[171, 336] Further, there is widespread distrust in official results and water quality in the aftermath of

the Flint water crisis, causing some consumers to avoid tap water use and/or test their own water using off-the-shelf kits that are not reliable Standard Methods.[72, 263, 280] These gaps in the LCR highlight the need for guidance to help residents and proactive utilities address lead and copper corrosion issues.

1.1.3 Addressing Lead and Copper Corrosion in Drinking Water

Although corrosion chemistry is complex, the extent of lead and copper release from plumbing materials into drinking water generally depends on several factors related to water chemistry. Lead and copper in contact with drinking water can transition over time to lower solubility mineral solids, or "protective scales," inside plumbing.[44, 106, 114, 155, 179, 185, 262, 303, 304, 339] The formation and stability of these protective scales depends on water chemistry and, in the case of copper, can take anywhere from minutes to decades or more.[44, 113, 155, 262, 302, 304, 347] The two standard corrosion control treatments (CCT), increasing the water pH or adding corrosion inhibitors such as orthophosphate, can reduce lead and copper release by facilitating the formation of protective scales or reducing scale dissolution.[98, 262, 304] However, continuous CCT may be required to maintain lead and copper reductions and different doses may be required control lead vs. copper.[44, 302, 304]

Because of the LCR's preference for detecting and addressing lead problems, utilities may not detect and be expressly required to implement CCT to control elevated cuprosolvency. Therefore, additional guidance is needed to help utilities address cuprosolvency issues. The NDWAC Consensus Statement proposed that many waters could be classified as "non-aggressive" to new copper pipes based on key water quality parameters ("minimum" pH or orthophosphate dose) as a function of alkalinity. Utilities meeting such criteria could be relatively assured that very few homes would have copper above the 1.3 mg/L AL after a few

months of exposure to the water, and could, therefore, potentially be exempted from burdensome verification sampling for copper if it were included in the LCR.[15] While the most recent LCR revisions did not follow these recommendations they could still serve as "best practice" guidance for building managers, residents, or utilities who still wish to address these concerns on a voluntary basis. Chapters 3 and 4 build upon these recommendations by experimentally determining the "minimum" orthophosphate doses or pH values needed for given alkalinities to constitute effective cuprosolvency control and demonstrate several challenges in broadly applying this approach.

Low-cost at-home tests are commercially available and could help residents detect lead and copper issues. Residents may turn to these tests for many reasons, including greater mistrust in official results as a repercussion of the Flint Water Crisis.[263, 280] However, there is no certification authority or systematic evaluations, to determine the efficacy of at-home water tests, so their accuracy and reliability are uncertain. Chapter 2 systematically explored the accuracy and precision of commercially available at-home tests to help residents detect lead in drinking water.

Generally, avoidance strategies such as flushing water before use, using certified point of use filters, or using bottled water are suggested if elevated lead and copper are detected in drinking water. However more costly approaches such as replacing lead or copper bearing plumbing may also be suggested.[262] It is possible that short-term site specific point of entry treatments could facilitate permanent formation of low-solubility protective scales.[44] However, permanent water chemistry adjustments, analogous to utility CCT treatment (i.e., dosing phosphate, raising pH), may be needed and the long-term costs and maintenance considered. In Chapter 5, at-home tests for copper and pH are considered to support copper detection and selection of single-building treatments to help residents address cuprosolvency issues.

1.2 Overview and Attribution

Chapter 2 describes the accuracy and challenges surrounding use of at-home test kits to measure lead in drinking water. A survey of Amazon.com was conducted to identify and procure available residential level lead in water test kits. The accuracy and ability of these kits to indicate dissolved and particulate lead at concentrations relevant to drinking water is discussed. Finally, an at-home dissolution method using lemon juice or vinegar is considered to facilitate detection of lead from particles. This work, titled "Challenges of detecting lead in drinking water using at-home test kits", was published in *Environmental Science and Technology* in January of 2021. This work was co-authored by Dr. Kelsey J. Pieper (Conceptualization, Methodology, Supervision, Review and editing, Funding acquisition), Dr. Jeffrey Parks (Methodology, Review and editing), and Dr. Marc Edwards (Conceptualization, Methodology, Supervision, Review and editing, Funding acquisition).

In Chapter 3 a guidance framework is considered to help utilities address cuprosolvency concerns in drinking water via orthophosphate corrosion control treatment. In support of this framework, the "minimum" orthophosphate doses to yield copper below the 1.3 mg/L EPA action level are determined for given alkalinities via laboratory cuprosolvency tests using copper tube segments. Finally, challenges applying cuprosolvency test methods based on fresh cupric solids are demonstrated. The manuscript "Considering a utility-centric framework for mitigation of elevated cuprosolvency in drinking water" was submitted to *Environmental Science and Technology* in April of 2023. It was co-authored by Emily Smith (Experimental support, Review and editing), Grace Byrd (Experimental support, Review and editing), Dr. Michael Schock (Methodology, Review and editing), and Dr. Marc Edwards (Conceptualization, Methodology, Supervision, Review and editing, Funding acquisition).

Chapter 4 presents guidance to help utilities address cuprosolvency concerns using pH-based corrosion control. The "minimum" pH values needed to yield copper below the 1.3 mg/L EPA action level are determined for given alkalinities in copper tube cuprosolvency tests. Further, several challenges are discussed that could complicate broad application of pH corrosion control based on these simple criteria. Results indicate that factors such as tube manufacturer, aging, and water quality parameters like natural organic matter, sulfate, and silica concentrations warrant further consideration. This paper, titled "Challenges predicting cuprosolvency in drinking water based on common water chemistry parameters," will be submitted to *Environmental Science and Technology*. It was co-authored by Dr. Marc Edwards (Conceptualization, Methodology, Supervision, Review and editing, Funding acquisition).

In Chapter 5, guidance previously developed for utilities is adapted to suit residential needs for addressing cuprosolvency issues. At-home tests and water treatments are evaluated to determine their utility as tools to help residents detect and address cuprosolvency issues. Further, two illustrative case studies are presented to demonstrate use of the residential interventions put forth in the framework. A manuscript titled "Guidance for Residents Addressing Copper Problems in Drinking Water: Opportunities and Challenges" will be submitted to *Environmental Science and Technology*. Dr. Marc Edwards co-authored this paper (Conceptualization, Methodology, Review and editing, Supervision, Funding acquisition).

Chapter 6 presents the novel contributions and conclusions of this work and outlines the future research needed to better understand and more broadly apply these results to address utility and residential corrosion issues in drinking water.

Additional manuscripts that contribute to my body of work are presented in Appendices E to I. Appendix E presents a high-level analysis of sources of lead in drinking water, key variables affecting lead release, and potential remediation/avoidance strategies to address lead issues. This work, titled: "Understanding lead in water and avoidance strategies: a

United States perspective for informed decision-making" was published in *Journal of Water and Health* in 2019. This work was co-authored by Dr. Kelsey J. Pieper (Conceptualization, Methodology, Writing, Funding acquisition), Dr. Adrienne Katner (Conceptualization, Methodology, Writing, Funding acquisition), Dr. Min Tang (Methodology, Review and editing), and Dr. Marc Edwards (Conceptualization, Methodology, Review and editing, Funding acquisition). Appendix G identifies and attempts to apply various chromium VI groundwater source tracking techniques to evaluate coal ash contamination in North Carolina well water via a large citizen science sampling campaign. This work, titled "Addressing emerging contaminants in well water: occurrence of hexavalent chromium in North Carolina," will be submitted in summer of 2023. This work was co-authored by Dr. Kory Wait (Well sampling and recruitment, Data and statistical analysis, Review and editing), Nathan Jones (Data and statistical analysis, Review and editing), Dr. Andrew George (Methodology, Well sampling and recruitment, Review and editing), Wilson Mize (Methodology, Well sampling and recruitment, Review and editing), Brady Freeman (Methodology, Well sampling and recruitment, Review and editing), Jefferson Currie (Methodology, Well sampling and recruitment, Review and editing), Dr. Michael Burchell (Methodology, Well sampling and recruitment, Review and editing), Dr. Jeffrey Parks (Sample and data analysis), Dr. Adrienne Katner (Methodology, Review and editing), Dr. Marc Edwards (Review and editing, Funding acquisition), and Dr. Kelsey Pieper (Methodology, Supervision, Well sampling and recruitment, Review and editing, Funding acquisition). Finally, Appendix I presents a discussion of challenges academics may face engaging in citizen science via a collection of case studies focused on addressing water safety issues. This manuscript, titled "Experiences in Citizen Science Related to Consumer Perceptions of Water Safety (2011-2022)", will be submitted to *Environmental Science and Technology Water*. It is co-authored by Dr. Kory Wait (Methodology, Writing and compiling, Case study lead, Review and editing), Dr. Siddhartha Roy (Methodology, Supervision, Case study lead, Review and editing), Dr. Adrienne Katner

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Chapter 2

Challenges of Detecting Lead in Drinking Water Using at-Home Test Kits

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Abstract

Lead in drinking water remains a significant human health risk. At-home lead in water test kits could provide consumers with a convenient and affordable option to evaluate this risk, but their accuracy and reliability is uncertain. This study examined the ability of at-home lead test kits to detect varying concentrations of dissolved and particulate lead in drinking water. Sixteen brands representing four test kit types (binary color, binary strip, colorimetric vial, and color strip) were identified. Most kits (12 of 16 brands) were not suitable for drinking water analysis, with lead detection limits of 5−20 mg/L. Binary strips detected dissolved lead at drinking water-relevant levels but failed to detect particulate lead. Household acids (lemon juice and vinegar) improved the strip's ability to detect lead by dissolving some of the lead particulates to the point soluble lead exceeded 15 µg/L. These results illustrate the applications of at-home testing kits for drinking water analysis, highlight limitations and areas for possible improvement, and put forth a testing protocol by which new at-home lead test kits can be judged.

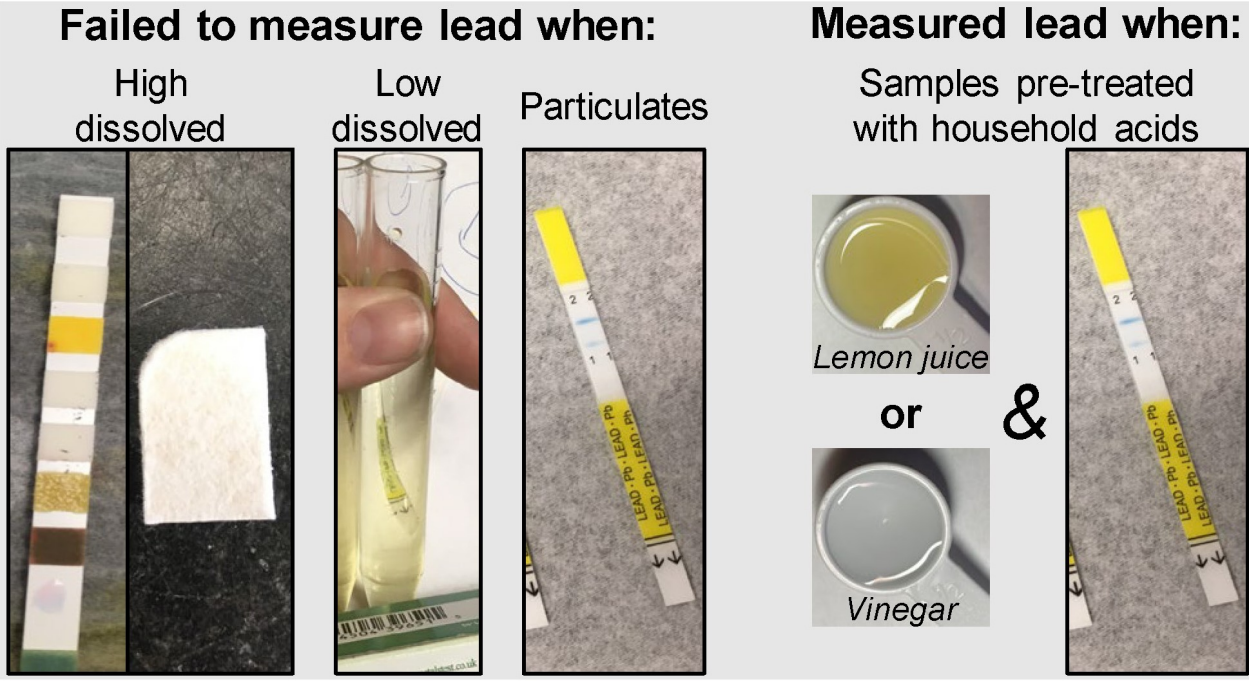


Figure 2.1: Table of Contents Art

2.1 Introduction

Lead in drinking water remains a significant human health risk, as evidenced by recent cases of water-related elevated blood lead[55, 112, 117, 142] and thousands of community systems with Lead and Copper Rule (LCR) violations.[239] There are well-known weaknesses in the protection offered to individual homes under the LCR. Weaknesses include improper sampling methods and analytical procedures that can miss water lead hazards, as well as the provision that allows up to 10% of the high risk homes tested to have very high water lead without triggering public notification.[171, 342] Although problems with high lead at a given tap are frequently viewed as government or regulatory failures, it is important to note that the LCR was not designed to protect every consumer at every tap—rather, it aims to reduce the magnitude and frequency of lead in water problems systemwide.[336, 342, 344]

This leaves residents with a share of the responsibility to identify and address their lead in water problems.

Portable and consumer-centric tools are urgently needed to enable residents to meet that responsibility, but there is an important balance between convenience and data quality that must be understood.[334] On-site testing using spectrophoto-metric, fluorescence-based methods and anodic stripping voltammetry (ASV) analysis has been explored because of their portability, fast results, relative ease of use, and low costs.[63, 100, 334] However, these tests may be subject to interferences from other analytes in water and may not detect particulate lead without adding an acidification step, thereby underestimating total lead in drinking water.[63, 100, 171, 334] While these technologies have lower capital costs than laboratory testing, costs can still be significant when factoring in the price per sample (about $10 plus labor cost) and the portable ASV unit cost (about $1000–2000).[100, 334] Moreover, the US Environmental Protection Agency (US EPA) recommends that users exercise caution when choosing a field testing technology, as some analyzers use dangerous consumables and reagents, and require technical skills. This makes them better suited for researchers and utilities, so they are not recommended or are poorly suited for use by residents.[63, 300, 334] In contrast, low-cost commercially available at-home lead in water test kits may provide a good alternative for consumers; however, there has been limited research into their efficacy, so their accuracy and reliability are uncertain. Several organizations (e.g., Good Housekeeping Institute, Consumer Reports) have evaluated these commercially available at-home kits, but these are not systematic evaluations and they failed to evaluate the expected problems associated with detecting particulate lead.[11, 93]

Distrust of drinking water quality, which increased in the aftermath of the Flint water crisis, has resulted in tap water avoidance which has significant health and economic implications.[29, 263, 280] The use of a low-cost, accurate at-home lead in water test kit could increase the

confidence in tap water quality and help address water insecurity in at-risk communities. This study explored the accuracy and precision of commercially available at-home lead in drinking water test kits when exposed to varying water conditions. Specifically, the objectives of this study were to (1) identify available at-home test kits that measured lead in water; (2) examine the ability of test kits to detect different concentrations of dissolved and particulate lead in water; and (3) characterize the ability of test kits to detect solder particles, reported to be relatively recalcitrant,[90] exposed to water with different corrosion control treatments. This is the first scientific study assessing the accuracy and precision of a range of available at-home lead in water test kits that are commercially available and in use despite the lack of scientific validation or certification. Moreover, as new test kits become available, the testing protocol outlined in this study can serve as a metric to ensure that the kits can accurately detect lead in drinking water.

2.2 Methods

2.2.1 Test Kit Selection

An Amazon search for "water lead test kit" was performed on May 9, 2018. This search yielded 347 products, of which 122 (35.1%) measured lead in water(Figure A.1). After removing duplicates, 34 unique test kits were identified. This study examined at-home test kits, so spectrophotometric (n = 1) and mail-away tests (n = 8) were removed. The remaining test kits were categorized into four groups (Figure A.2): (1) binary strips (n =8), which indicate lead detection based on the appearance of lines, and color-based tests including: (2) colorimetric vial tests (n = 1), (3) binary color tests (n = 1), and (4) color strips (n =15), which indicated lead detection or concentration based on color changes.

Four brands of color strips would have been included but were not available at the time of purchase. Upon purchase, we discovered that seven of the binary strip brands had identical interior packaging (5 of one type and 2 of another type). This suggested that the same testing materials were being used among several brands; therefore, three brands were chosen to represent the three unique binary strip testing materials identified. The remaining 16 test kits were evaluated: 3 binary strip, 1 colorimetric vial test, 1 binary color test, and 11 color strip brands. The detection levels of these test kits were 15, 20, 5000, and 20,000 µg/L, respectively.

2.2.2 Testing Approach

Testing was performed according to the manufacturers' instructions. Each brand was tested in triplicate, and one blank control was included for each water condition per phase (Figure 2.2). The base water condition was a modified version of the NSF/ANSI 53 pH 8.5 water,[72] with dissolved lead added as $Pb(NO_3)_2$ and particulate lead as described below. Testing was conducted within 1 h of adding dissolved lead and 30 min of adding particulate lead. To determine the accuracy and precision of the test kits, a tiered testing approach was developed. Specifically, the test kits were exposed to high dissolved lead (Phase 1) and, based on their performance, were exposed to low dissolved lead (Phase 2A) or solder particles (Phase 2B). Kits that were successful in Phase 2A were tested in Phases 3–5 with particulate lead. Six participants were recruited to read measurements for the high dissolved lead experiments (Phase 1). Results for lead concentrations were calculated based on the average of the triplicate tests for each of the participants. Only one participant read results for the other experiments (Phases 2–5), which focused primarily on the binary strips. Overall, participants were not confident in their ability to read the lead measurements (Section A.1), but the binary strips had consistent readings among participants based on the presence/absence

metric.

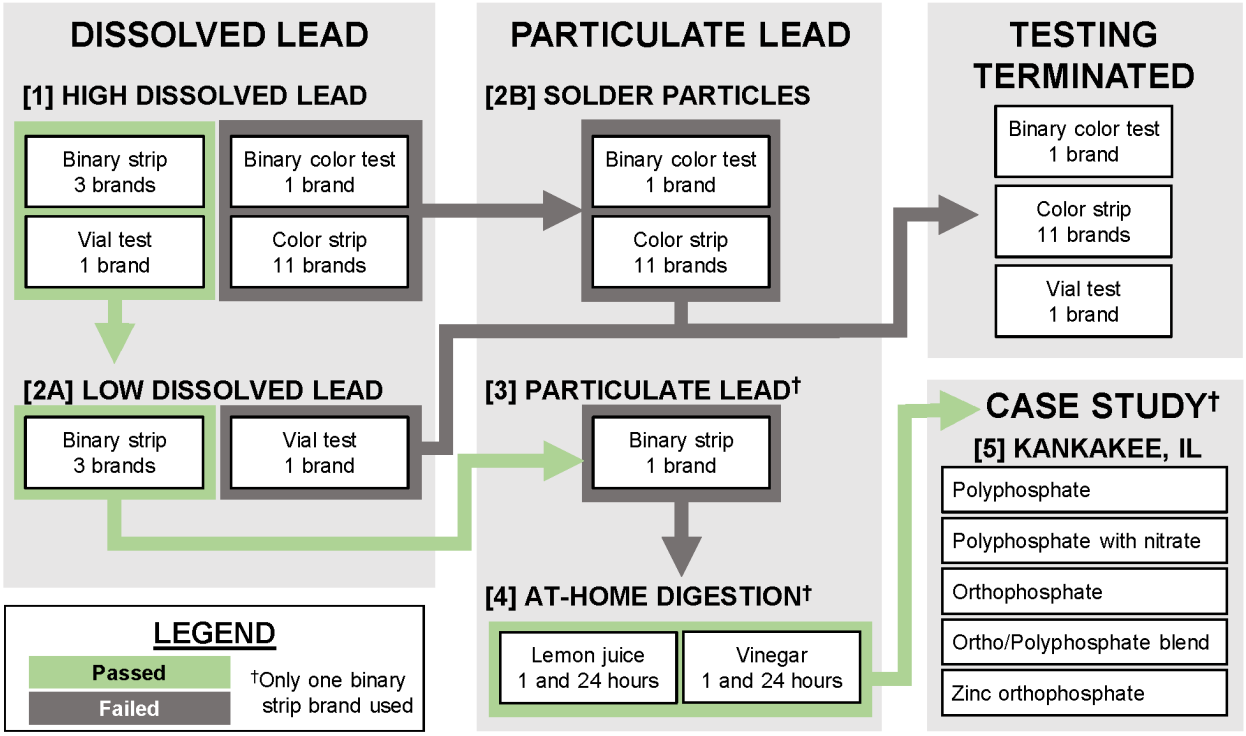


Figure 2.2: **Phases of lead in water testing used to evaluate test kits' ability to detect or measure dissolved and particulate lead.**

Dissolved Lead

All 16 test kits were evaluated in Phase 1 to determine if they could measure or detect a relatively high dissolved lead concentration (150 µg/L). The four test kits (1 colorimetric vial test and 3 binary strip brands) that accurately characterized high dissolved lead were then evaluated in Phase 2A to determine if they could detect low dissolved lead (5, 10, and 20 µg/L) near the US EPA action level.

Particulate Lead

The 12 test kits that did not accurately characterize high dissolved lead in Phase 1 (1 binary color and 11 color strip test brands) were evaluated in Phase 2B to determine if they could measure or detect an extremely high level of particulate lead, which was well above their detection limits. Lead solder particles were added to the base water to produce a lead concentration of 50 mg/L.

Three binary strip brands detected low dissolved lead in Phase 2A, but only one brand (Brand 1) was selected for testing in Phases 3 and 4, to determine if the binary strips could detect various types of particulate lead in water. Brand 1 was selected for this additional testing because it consistently detected lead at 10 µg/L.

All particle samples, except for the particles from Cicero, IL (Fe-PO_4), were collected as dry particles from pipe surfaces and added (5.0 ± 0.8 mg) to 1 L of the base water amended with 1 mg/L phosphate as P to reduce dissolution in water. The Cicero, IL particles were tested in the municipal tap water in which they were collected, which had a phosphate concentration of 0.31 mg/L as P. Representative lead particles (range of % lead content of total recovered particle mass from n = 3–4 samples; particle name) were derived from (1) lead pipe from a laboratory experiment (92-99% Pb; Pb);[214] (2) a solder joint extracted from a Flint, MI home (42-70% Pb; Pb–Sn); (3) lead pipe from Washington, DC (55-67% Pb; Pb–Fe); (4) lead pipe from Buffalo, NY (23-30% Pb; Al–Pb-1); (5) lead pipe from Flint, MI (9-27% Pb; Al–Pb-2); (6) a water sample from Cicero, IL (3-14% Pb; Fe-PO_4); and (7) a galvanized iron pipe from resident zero in Flint, MI (0.8-1.4% Pb; Zn–Fe)[236] (Table A.2; Figure 2.3). The total recovered particle masses across all samples were 24–97% of the total particle mass added, with the remainder attributable to incomplete particle dissolution and/or mass from constituents such as oxygen and carbon that were not detected (Table A.3). To illustrate

the variable composition across particle types, Figure 2.3 presents the composition and mass recovery for one of the 3–4 samples per particle type used in this analysis. Section A.2 provides an overview of particle characterization to determine lead content.

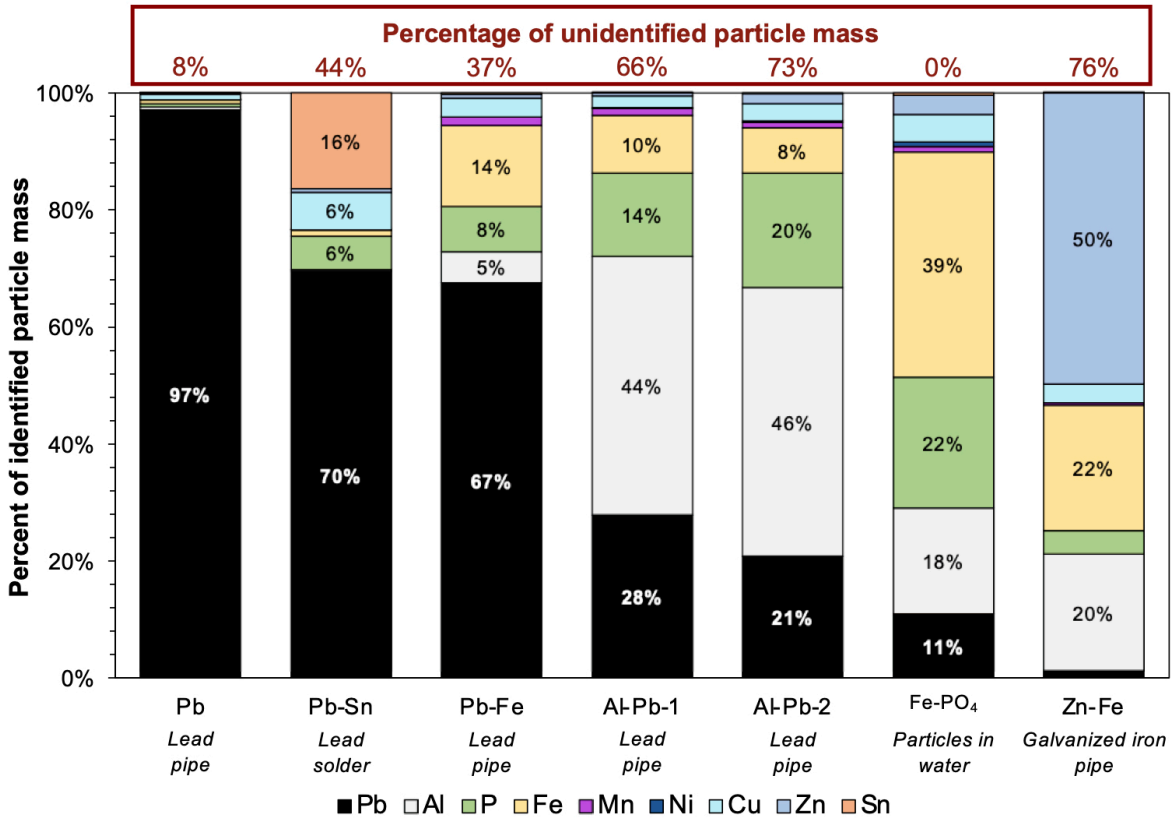


Figure 2.3: **Metal composition for one particle sample of each type** (5 mg dry particles added to 1L of water or particles collected in water) based on the total recovered particle mass. Data labels are not included for metals that represent <5% of the particle composition.

Particle size distributions were determined for dry particles via Raman atomic force microscopy (WITec AFM-Raman). Images at 4× and 40× magnifications were analyzed using ImageJ software (National Institute of Health) to count particles of varying sizes, with a threshold of four pixels to reduce image noise resulting from color threshold adjustment.

Representative images for dry particles from Phases 2B and 3 are presented in Figure A.3.

Since the binary strip could not detect lead particles, experiments were conducted to determine if common household acids (i.e., vinegar and lemon juice) could improve detection by dissolving lead particles in Phase 4. A dose of 41 mL of lemon juice or vinegar per 1 L of the water condition was added and digestion occurred for 1 h and 24 h.

Case Study

Lead solder particles are sometimes more difficult to dissolve than other lead particles.[90] Therefore, lead particles were formed by exposing lead solder to a base water with different types of phosphate inhibitors, and binary strip detection was evaluated with and without at-home dissolution (Phase 5). Finished drinking water from the Kankakee River (Table A.4) was used to create a more realistic relationship between dissolved and particulate lead observed in drinking water. In brief, finished drinking water was shipped to Virginia Tech, where it was blended with synthetic water in a 2:1 ratio and the pH was adjusted to 8.2. Incorporating the synthetic water increased the chloride to sulfate mass ratio to 0.76, which created a water condition scenario with higher potential for galvanic corrosion.[112] Lead solder joints were exposed to the blended water with five CCT strategies: (1) polyphosphate (0.8 mg/L as P); (2) polyphosphate with nitrate (0.8 mg/L polyphosphate as P, 3 mg/L nitrate); (3) orthophosphate (1 mg/L as P); (4) 90:10 orthophosphate/polyphosphate blend (1 mg/L as P); and (5) zinc orthophosphate (0.5 mg/L Zn and 1 mg/L orthophosphate as P). In addition, an amended control (blended water with no inhibitors) and an unamended control (finished drinking water with no inhibitors) were analyzed. Prior to sampling, all coupons were conditioned using a daily dump-and-fill protocol for (1) four days with pH 8 water with 2 mg/L chlorine as Cl_2 and an alkalinity of 500 mg/L as $CaCO_3$ from sodium bicarbonate, (2) 12 days of conditioning using finished drinking water, and (3) 36 days of

conditioning using their water condition. After conditioning, a composite water sample was collected from each water condition (n = 5 coupons per condition, n = 10 coupons per control) for testing.

Validity of Binary Strip Test Kits.

Sensitivity, specificity, and accuracy were calculated for all binary strip tests. Sensitivity represented the number of correctly identified positive lead results (i.e., $\geq$ 15 µg/L) divided by the true number of positive results (true positive + false negative). Specificity represented the number of correctly identified negative lead results (i.e., <15 µg/L) divided by the true number of negative lead results (true negative + false positive). Accuracy represented the total number of correctly identified lead results divided by the total number of tests. In total, 141 tests were conducted in Phases 1−5, with 113 tests of Brand 1 and 14 tests of Brands 2 and 3 each. Seven kits were removed because of inconclusive readings. These parameters were calculated for tests exposed to only dissolved lead (Phases 1 and 2A), only particulate lead (Phase 3), and both dissolved and particulate lead (Phases 4 and 5). Results were then calculated for all binary tests (n = 134) considering only dissolved lead concentrations and then total lead concentrations.

2.2.3 Analytical Methods

Base water parameters and chlorine were analyzed using standard methods.[72] To determine the concentrations of dissolved and particulate lead, a 10 mL filtered (0.45 µm) aliquot (i.e., dissolved lead) and total lead sample (i.e., digestion in the bottle to measure all recovered lead) were analyzed. Particulate lead was defined as the difference between total lead and dissolved lead concentrations. Metal analyses of dissolved lead were performed

after filtration and acidification with 2% nitric acid for at least 16 h. Total recoverable particle mass was defined as the sum of the masses of eight metals and phosphorous (Table A.3) in fully digested samples (Section A.2). Metal analyses for total lead and total particle mass recovered were performed after acidification in the sample bottle with 2% nitric acid and 2% hydroxylamine and heated to 60−65 °C in an oven for at least one week prior to analysis.[258] Phosphorus and metals were measured using ICP−MS (Thermo Scientific iCAP RQ ICP−MS) via method 3125 B.[72] QA/QC was performed after every 10 samples.

2.3 Results

2.3.1 Detecting Dissolved Lead.

Phase 1: Detecting High Dissolved Lead.

When measuring high dissolved lead in water (150 µg/L), users reported inaccurate lead concentrations for 12 of the 16 kits (11 color strip brands and 1 binary color test brand) (Figures 2.4 and A.4; Tables A.5 and A.6). Users reported no lead in water with three of the color strip brands, as expected, because the dissolved lead concentration was below the lowest measurement increment of 20,000 µg/L. However, users reported substantially more dissolved lead than was present with 8 of the 11 color strip brands, with average reported concentrations of 1700−45,000 µg/L (11−300 times higher than the actual value). Users also reported lead in the blank samples for 6 of these 8 strips (mean: 16,000 µg/L, standard deviation (SD): 18,000 µg/L). The binary color strip did not detect lead in water, which was expected because its detection limit was 5000 µg/L. These 12 kits did not accurately measure or detect levels of lead considered high in drinking water (10× the US EPA action level), making them poorly applicable for drinking water testing. Because of their high detection

limits, these kits were evaluated once again when exposed to extremely high particulate lead concentrations in Phase 2B (Figure 2.2).

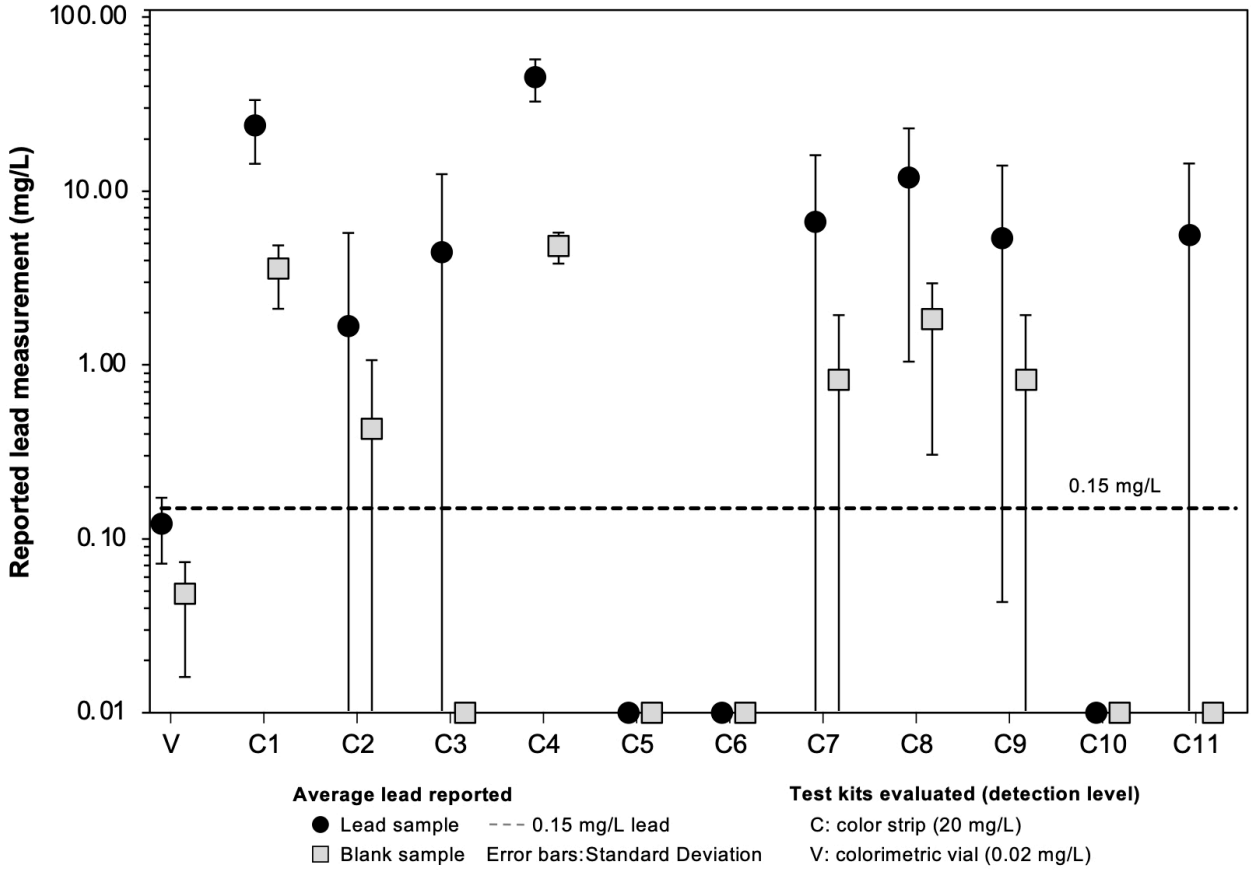


Figure 2.4: **Average lead reported by participants for at-home tests** with numeric results (1 colorimetric vial test and 11 brands of color strips) when measuring (1) 150 µg/L dissolved lead (black circles) (n = 6) and (2) blank (grey squares) (n = 6). Error bars represent standard deviations of averaged results from participants.

Users reported detection of lead and more accurate lead concentrations with the four remaining test kits (1 colorimetric vial test and 3 binary strips), with an average lead concentration of 122 µg/L (SD: 50 µg/L) when using the colorimetric vial test. However, an average lead level of 85 µg/L (SD: 66 µg/L) was reported in the blank, suggesting that users had problems distinguishing lead levels. The three binary strips detected lead when present, which was

expected since lead concentrations were 10 times their detection threshold of 15 µg/L. As these kits accurately measured and detected high dissolved lead, they were evaluated for detection of lower dissolved lead in Phase 2A (Figure 2.2).

Phase 2A: Detecting Low Dissolved Lead.

All binary strip brands detected lead in water when exposed to 20 µg/L (Figure 2.5), which was above the manufacturers' reported 15 µg/L detection limit. Several replicates among the brands also detected lead at 10 and 5 µg/L, highlighting some issues with their precision at low lead levels. The participant could not accurately measure low dissolved lead with the vial test, since reported measurements overestimated lead at each concentration (Figure 2.5). With the vial test, the participant reported an average lead concentration of 100 g/L for lead levels of 5−20 µg/L, including the blank. Although only one participant recorded measurements, the inability to read low lead levels was in keeping with the results from Phase 1, where six participants measured an average of 85 µg/L in the blank. As the participant could not accurately measure low dissolved lead, testing of the vial test was not continued. Since all binary strips performed similarly (i.e., detected low dissolved lead above their threshold), only testing of Brand 1 was continued in Phase 3 (Figure 2.2).

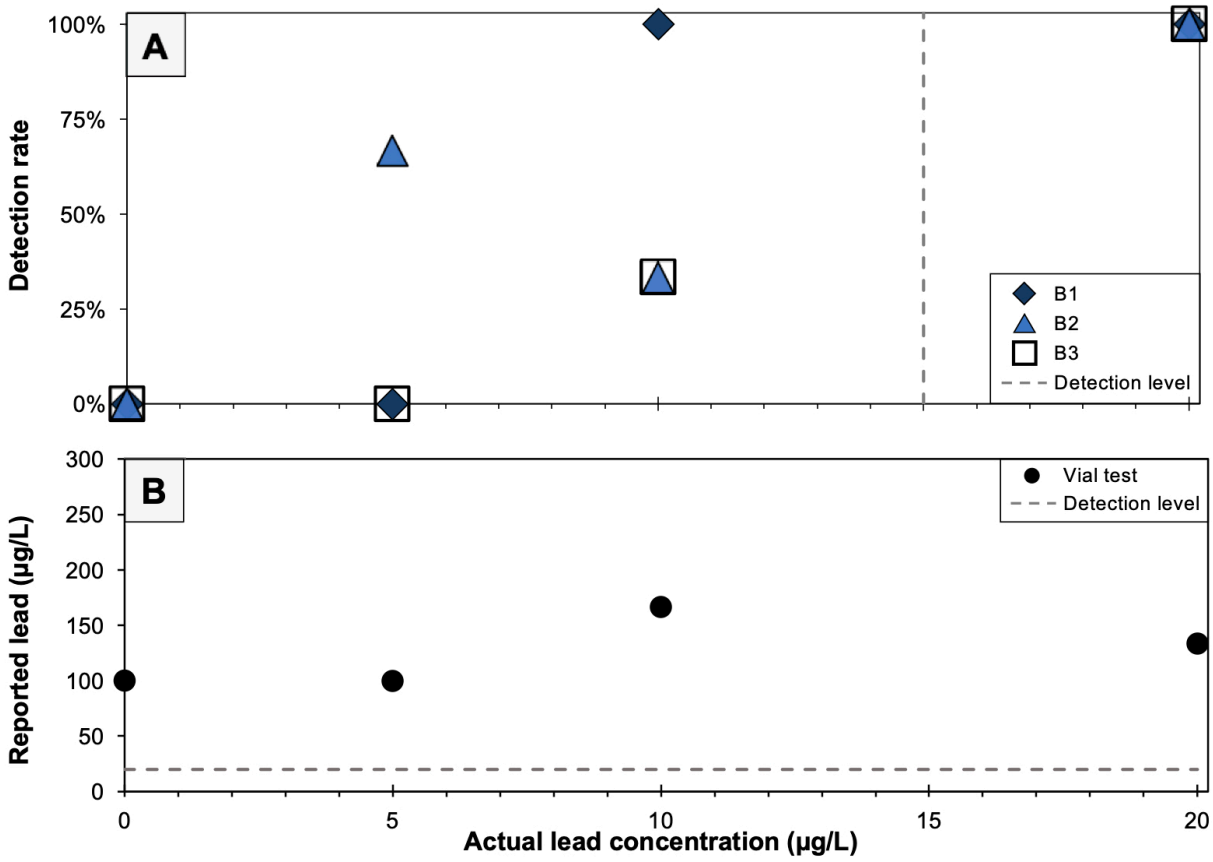


Figure 2.5: **Test kit detection of low dissolved lead** (0, 5, 10, and 20 µg/L) (n = 1 participant): (A) detection rate of binary strips (positive results/number of tests) and (B) average lead measurements from vial tests.

2.3.2 Detection of Particulate Lead.

Phase 2B: Detecting Extreme Concentrations of Lead Solder Particulates.

None of the color strips or binary color tests could reliably detect the presence of very high particulate lead (50 mg/L). The lead concentration was purposefully set very high, so it was well above all reported method detection thresholds of 5 and 20 mg/L. These results confirm the expectations that these kits cannot measure particulate lead. Only one replicate of the

color strip detected particulate lead and that was because a lead solder particle randomly stuck to the reactive surface of the strip (Figure A.4). As color strips and binary color test brands could not measure either dissolved or particulate lead at even very high concentrations for drinking water, testing was not continued (Figure 2.2).

Phase 3: Detecting Various Types of Lead Particulates.

A wide array of representative water lead particles was tested with one brand of binary strip in triplicate. Despite total lead concentrations above 15 µg/L (15−4392 µg/L; Tables 2.1 and A.3), the binary strips only detected lead in 2 of the 7 lead particle samples (Pb−Sn and Pb−Fe). In both of these cases, the dissolved lead concentrations were above the detection threshold (15 and 33 µg/L, respectively). The binary strips failed to detect lead when exposed to the other five samples, which all had dissolved lead below the detection threshold (0.4−2.8 µg/L). Similar to the test kits tested in Phase 2B (Figure 2.2), the binary strips were unable to detect particulate lead in water.

Table 2.1: **Lead Test Kit Results**
[a] % of total recovered particle mass could not be calculated for Fe-PO_4 particles because they were collected in water. [b] % increase in dissolution was not calculated when no treatment was performed since this constitutes the baseline for this calculation. Dashes indicate when test kit testing was not conducted. *Tested in triplicate, one test was invalid and not included in analysis. Pb: lead; Al: aluminum; Fe: iron; Zn: zinc; Sn: tin; PO_4: phosphate. Bold text denotes dissolved lead concentrations >15 µg/L.

Treatment	Contact Time (hour)	Particle Type	% of Total Recovered Particle Mass	Total Lead (µg/L)	Dissolved Lead (µg/L)	% Dissolved Lead of Total Lead	% Improvement in Dissolution (Treatment - None)	# Positive Test Strip Results/Total
None	< 0.5	Pb	92%	4391.5	1.3	0%	-	0/3
		Al-Pb-1	34%	495.9	0.5	0%	-	0/3
		Zn-Fe	24%	15.0	0.4	3%	-	0/3
		Fe-PO4	-	37.3	2.8	8%	-	0/3
		Pb-Sn	56%	2041.8	**33.7**	2%	-	3/3
		Al-Pb-2	27%	292.0	0.4	0%	-	0/3
		Pb-Fe	63%	2302.5	**15.5**	1%	-	3/3
Lemon Juice	1	Pb	97%	4466.9	**612.7**	14%	14%	3/3
		Al-Pb-1	43%	501.1	**70.0**	14%	13%	2/3
		Zn-Fe	35%	14.4	3.0	21%	18%	-
		Fe-PO4	-	35.7	**34.0**	95%	88%	2/2*
		Pb-Sn	58%	1781.3	**1460.2**	82%	80%	3/3
		Al-Pb-2	45%	228.1	**36.5**	16%	16%	3/3
		Pb-Fe	79%	2278.2	**869.6**	38%	37%	3/3
	24	Pb	97%	4466.9	**2782.3**	62%	62%	-
		Al-Pb-1	43%	501.1	**385.2**	77%	77%	-
		Zn-Fe	35%	14.4	13.0	90%	87%	-
		Fe-PO4	-	35.7	**35.4**	99%	92%	-
		Pb-Sn	58%	1781.3	**1698.9**	95%	93%	-
		Al-Pb-2	45%	228.1	**172.9**	76%	76%	-
		Pb-Fe	79%	2278.2	**1770.2**	78%	77%	-
Vinegar	1	Pb	88%	4733.4	**435.1**	9%	9%	3/3
		Al-Pb-1	39%	591.0	**73.4**	12%	12%	3/3
		Zn-Fe	25%	17.1	0.9	5%	2%	-
		Fe-PO4	-	34.4	**31.7**	92%	85%	2/2*
		Pb-Sn	27%	598.9	**423.8**	71%	69%	3/3
		Al-Pb-2	31%	492.6	**29.8**	6%	6%	2/2*
		Pb-Fe	63%	1920.4	**492.7**	26%	25%	2/2*
	24	Pb	88%	4733.4	**1111.3**	24%	24%	-
		Al-Pb-1	39%	591.0	**308.0**	52%	52%	-
		Zn-Fe	25%	17.1	4.9	29%	26%	-
		Fe-PO4	-	34.4	**31.4**	91%	84%	-
		Pb-Sn	27%	598.9	**493.8**	83%	81%	-
		Al-Pb-2	31%	492.6	**201.3**	41%	41%	-
		Pb-Fe	63%	1920.4	**926.5**	48%	47%	-

2.3.3 At-Home Dissolution of Particles.

Lemon Juice Dissolution.

Potential dissolution of lead particles using common at-home acids was explored to improve the accuracy of binary strips. Dissolution using lemon juice for 1 h (Tables 2.1 and A.3) had mixed results, with dissolved lead representing 14−95% of the total lead. Almost all of the total lead in the Fe-PO_4 particles (95%) dissolved after 1 h of lemon juice digestion. However, limited lead dissolution (<21% of the total lead) was observed with Pb, Al−Pb-1, Zn−Fe, and Al−Pb-2 particles. When considering the particle size (Tables A.7 and A.8), three particle conditions (Al−Pb-1, Al−Pb-2, and Zn−Fe) had relatively larger particles, with >47% of the detected particles having diameters >10 µm. The other four particle conditions (Pb, Pb−Sn, Pb−Fe, and lead solder) had smaller particles, with >91% of the particles having diameters <10 µm. Conditions that initially had larger particles represent three of the four conditions with limited lead dissolution, whereas greater lead dissolution was found in most conditions with smaller particles, as per expectations.[286] When dissolved lead levels increased above the 15 µg/L detection threshold, test kits correctly detected the presence of elevated lead in water. Dissolution using lemon juice facilitated the detection of five previously undetected lead particles in this study. Binary strip testing for the Zn−Fe particle was not conducted because the total lead concentration was less than 15 µg/L, but lemon juice digestion did increase dissolved lead levels by 20.8%. Increasing the lemon juice digestion time to 24 h further increased dissolved lead concentrations from an average (range) of 40% (14−95%) after 1 h to 83% (62−99%) after 24 h.

Vinegar Dissolution.

When exposed to vinegar for 1 h (Tables 2.1 and A.3), dissolved lead represented 5−92% of the total lead. As before, 92% of the total lead in Fe−PO4 particles dissolved, whereas <12% of the total lead in Pb, Al−Pb-1, Zn−Fe, and Al−Pb-2 particles dissolved after 1 h of vinegar dissolution. Dissolved lead concentrations increased from an average (range) of 32% (5−92%) after 1 h to an average of 52% (24−91%) after 24 h. Again, these results demonstrated that at-home treatment with vinegar improved the ability of the binary strip to detect particulate lead for five of the particulate conditions. However, dissolution with lemon juice consistently yielded greater lead dissolution compared with vinegar treatment, which is likely due to the lower pH achieved with lemon juice treatment (pH 3) compared with vinegar (pH 3.5).

2.3.4 Case Study: Detection of Lead Solder Particles Formed in Varying CCT Waters.

The accuracy and precision of binary strips were examined using lead particles generated when lead solder joints were exposed to five different corrosion control treatments (Tables A.9 and A.9). Binary strips detected lead in the controls, polyphosphate, and polyphosphate with nitrate conditions, which had dissolved lead above the detection threshold (130.2−443.6 µg/L). However, binary strips did not detect lead in the orthophosphate or orthophosphate/polyphosphate blend conditions because of the low dissolved lead (2.5 and 4.8 µg/L), despite the total lead being above the detection threshold (20.1 and 17.2 µg/L). Binary strips correctly indicated no lead in the zinc orthophosphate condition when the total lead was below 15 µg/L. Overall, binary strips detected lead in 4 of the 6 conditions where the total lead exceeded the detection threshold of 15 µg/L.

Lemon juice treatment increased the percent of total lead that was dissolved in controls, polyphosphate, and polyphosphate with nitrate condition (10−63 to 91−95%), thereby producing higher dissolved lead concentrations (185.5−530.3 µg/L). However, these conditions had high enough dissolved lead without digestion to facilitate binary strip detection. As expected, binary strips did not detect lead in the orthophosphate or orthophosphate/polyphosphate blend conditions where the total lead was below 15 µg/L. However, lead dissolution increased with lemon juice treatment from 13−28 up to 67% of the total lead, suggesting this treatment would improve detection with binary strips if high enough total lead was present. As before, the use of at-home digestion increased the dissolution of lead, which tended to improve the accuracy of the binary strips.

2.3.5 Binary Strip Accuracy.

Binary strips' inability to detect particulate lead resulted in lower sensitivity, specificity, and accuracy (Table A.11). Binary strips were 82.1% accurate when testing water with only dissolved lead (Phases 1 and 2A), but only 28.6% accurate when testing water with only particulate lead (Phase 3). When water contained both dissolved and particulate lead (Phases 4 and 5), the accuracy increased to 89.2%. Overall, in Phases 1−5 binary strips had a 77.6% accuracy rate for total lead detection and 93.3% for dissolved lead detection. Increases in sensitivity and specificity were also observed when comparing detection based on total lead versus dissolved lead (total lead: sensitivity 77.2%, specificity 78.8%; dissolved lead: sensitivity 97.5%, specificity 87.0%). These results further underscore the limitations of binary strips in detecting particulate lead.

The relatively high false negative rates observed in Phases 4 and 5 (10.8%) and overall (17.2%) may also be concerning because these indicate kits reporting no lead when total

lead concentrations were actually >15 µg/L. While a high false negative rate (71.4%) was expected for particulate lead (Phase 3), clearly such problems also extend to situations with a mixture of dissolved and particulate lead. Overall, binary strip sensitivity, specificity, and accuracy at measuring dissolved lead in this study were relatively high, but lower accuracy, sensitivity, and specificity should be expected in situations where particulate lead is more prevalent. Future efforts should focus on either reducing such errors or considering how this error affects usefulness of the tests.

2.4 Implications

2.4.1 Engaging, Equipping, and Empowering Consumers in the LCR's Shared Responsibility Framework.

The 1991 LCR was never designed to protect all residents from elevated lead in drinking water, but instead created a shared responsibility between consumers and their water suppliers for mitigating lead in water exposure.[280, 342, 344] Water utilities are responsible for water quality targets for "optimal" corrosion control, regulatory monitoring of a few homes, removal of utility-owned lead service lines, and public education. Consumers share some responsibilities since lead in drinking water partly results from lead in privately owned building plumbing. However, traditionally, implementation of the LCR has focused almost exclusively on water utilities' responsibilities, and such utility-centric frameworks cannot fully address issues with lead in drinking water without engaging consumers.[112, 117, 142, 236] Likewise, private water systems (e.g., wells, springs) are not regulated by the US EPA, and in these cases, the sole responsibility for the detection and mitigation of lead falls on consumers.[262] It is necessary to provide consumers with the tools and resources to identify lead in drinking

water risks when present and instructions for protective interventions.

The availability of affordable, reliable, and accurate lead in water test kits could help residents identify water hazards and, in the aftermath of Flint, there are many new tests under development. While many existing at-home lead in water test kits are available, most were ineffective at detecting or measuring lead at drinking water levels. Binary strips, especially when coupled with at-home weak acid dissolution, reliably detected dissolved lead concentrations above 15 µg/L, suggesting that these kits show a lot of promise. However, only detecting lead above the existing 15 µg/L detection threshold may render these kits quickly obsolete, as many organizations are pushing for lower recommended lead in water levels, such as the 1 µg/L goal of the American Association of Pediatrics which has influenced policy decisions, interventions, and consumer expectations.[256] The test methodology developed herein could be used as a framework for vetting the new water lead test kits by developers.

2.4.2 Particulate Lead is an Emerging Problem in Drinking Water.

Lead particles from solder and pipe scales are an important source of lead exposure in drinking water[90, 109, 249, 335] that was not well quantified by at-home test kits. This is problematic because numerous cases of lead poisoning have been linked to ingestion of lead particles[112, 117] and many lead problems in the US are being attributed to the mobilization of particles.[90] In some cases, the likely presence of lead particles may be associated with discoloration, as was demonstrated through the association of higher lead concentrations with discolored water because of high co-occurring levels of iron (e.g., Flint water crisis).[236, 258] However, iron and lead do not always co-occur in drinking water and, in many cases, lead particles are not visible to the naked eye (e.g., Washington, DC Lead Crisis),[117] underscoring the importance of testing. As with other testing procedures,[63, 100, 171] at-

home test kits in this study failed to detect particulate lead, yielding false negative results.

Digestion with acids is needed to dissolve lead particles,[171] and household acids promoted enough dissolution of some lead particles to improve the binary strip detection of particulate lead. In addition to particle composition, variations in particle dissolution and the fraction of total recoverable particle mass may be related to unaccounted for particle constituents, as well as variations in observed particle size and surface area.[286] As expected, in this study, smaller particles generally exhibited greater dissolution than larger particles. Some lead particles (e.g., lead(IV) oxides and lead phosphates) have been reported to be insoluble in weak acid, whereas others (e.g., lead(II) oxides) are reported to be more readily dissolved.[90, 216, 249] Lemon juice and vinegar have been used to dissolve rust and metals[260] and may improve dissolution by lowering the pH of the solution (pH 3−3.5). Although the mineralogical identity of particles was not determined, lead phosphates were likely present in some particles as evidenced by phosphate >20% of the identified particle mass. The use of either vinegar or lemon juice for 24 h increased dissolved lead (relative to the total lead) by 41−92% for these particles. The relative ease with which lead particles in this study dissolved in weak acids suggests that some particles previously thought to be relatively recalcitrant may be more susceptible to dissolution in weak organic acids than previously thought. Therefore, at-home dissolution procedures may represent a relatively safe method to improve binary strip test kit performance in detecting particulate lead, reducing one of the main problems of at-home lead in water test kits.

2.4.3 Strengths and Weaknesses of Binary Strips.

This study demonstrated the critical weaknesses of some test kits, such as overestimation of lead concentrations and detection limits that exceed a drinking water-relevant range for

a majority of color-based test kits. Binary strips correctly indicated whether dissolved lead concentrations were above the 15 µg/Lthreshold in 93% of the tests in this study, but failure to detect particulate lead was a weakness of the binary strips. At-home dissolution with lemon juice or vinegar dissolved many particles enough to increase dissolved lead concentrations to >15 µg/L, allowing for detection of these particles. These results demonstrate that binary strips may present a somewhat accurate and relatively low-cost option ($13–$40 per test) for at-home testing. Future development focused on reducing the cost burden of lead testing, as exemplified by a goal to reduce at-home arsenic testing costs to as low as $0.21 per test,[59] could increase accessibility.

Lead in water test kits, similar to at-home arsenic tests, could benefit from improved ease of use, safety, accuracy, improved detection limits, and clear endpoints for easy interpretation.[59, 154, 195] Kits should include information about stagnation protocols, sample volumes, and flow rates, which are known sources of error/variation for lead sampling[45, 249, 314] as well as any additional dissolution steps or updates on regulatory or public health changes that affect the interpretation of results. For example, studies of similar binary strips for atrazine indicated that binary strips may require improvements in instructional materials or training to allow citizen scientists to more accurately determine results.[62] There is currently no certification authority to ensure accuracy of at-home lead in water test kits. In light of this, the sensitivity, specificity, and accuracy of all test kits needs to be verified under real-world conditions.

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Chapter 3

Considering a Utility-Centric Framework for Mitigation of Elevated Cuprosolvency in Drinking Water

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Abstract

Gaps in the United States Environmental Protection Agency (US EPA) Lead and Copper Rule(LCR) leave some consumers and their pets vulnerable to high cuprosolvency drinking water. This study seeks to help proactive utilities who wish to mitigate cuprosolvency problems through the addition of orthophosphate corrosion inhibitors. The minimum doses of orthophosphate necessary to achieve acceptable cuprosolvency in relatively new copper pipe were estimated as a function of alkalinity via linear regressions ($R^2 > 0.98$). Orthophosphate was very effective at reducing cuprosolvency in the short-term, but in some cases resulted in higher long-term copper concentrations than the corresponding condition without orthophosphate. Alternatives to predicting "long-term" results for copper tube using simpler bench tests starting with fresh $Cu(OH)_2$ solids showed some promise but would require further vetting to be effective.

3.1 Introduction

Cuprosolvency is the tendency of waters to cause release of soluble copper from pipes to water.[304] Elevated copper can cause aesthetic concerns (e.g. fixture staining) and both acute and long-term health concerns for consumers and their pets.[8, 115, 329] Cuprosolvency is one manifestation of copper corrosion, which can also include pinhole leaks(i.e. pitting), wall thinning, and erosion corrosion, all of which have unique physiochemical etiologies.[284] To reduce problems with elevated cuprosolvency throughout community water systems, the 1991 United States Environmental Protection Agency (EPA) Lead and Copper Rule (LCR) set an action level (AL) of 1.3 mg/L for the 90^{th} percentile copper concentration.[8, 106, 301, 344] However, the LCR only applies to municipal water systems and focuses sampling

efforts on older homes (pre-1986) at greatest risk for elevated lead—not on the homes with newer plumbing that is more likely to have elevated cuprosolvency.[44, 303, 339]

In the three decades since the LCR was implemented, the minimum age of the copper pipes in the pre-1986 sampling pool has increased from about 5 to 35 years, further exacerbating the disconnect between metal release from new copper and compliance monitoring results. Consequently, some utilities can have serious problems with metal release from new copper, without triggering any requirement for corrosion control based on LCR monitoring in older homes. Moreover, residents with private wells and other non-residential buildings are not regulated under the LCR.[49, 99, 304, 339] These gaps can leave private well owners and buildings with new plumbing in need of remedies for persistent high cuprosolvency.[44, 115, 270]

The 2015 National Drinking Water Advisory Council (NDWAC) consensus statement proposed updates to the LCR to address these deficiencies. The NDWAC asserted that many potable waters are inherently non-aggressive to new copper and utilities with such water should be exempted from burdensome LCR sampling.[15] To classify waters as "non-aggressive," water quality parameters meeting standards for low cuprosolvency should be defined for new plumbing.

It is generally thought that, as new copper pipes age in contact with drinking water, relatively soluble scales such as cupric hydroxide [$Cu(OH)_2$] initially form and control the maximum cuprosolvency in water.[304] Upon aging, these cupric hydroxide solids are thought to transition to less soluble solids (e.g., tenorite [CuO], malachite [$Cu_2(OH)_2CO_3$], and cupric phosphates).[44, 106, 114, 155, 179, 185, 303, 304, 339] Many water quality parameters such as oxidants, pH, chloride, sulfate, hardness, temperature, natural organic matter (NOM), microorganisms and nitrogen, sulfur, iron species, etc. may affect the rate of scale transitions through complex reactions including redox, complexation, crystallization, and crystal

poisoning.[113, 155, 302, 304, 347] It is believed that transitions from more soluble to less soluble solids can occur in timespans of minutes to decades, depending on water chemistry, and are irreversible in normal situations.[44]

The NDWAC proposed demarcating "non-aggressive" waters based on two standard corrosion control approaches at water utilities: dosing of orthophosphate inhibitors or adjusting pH/alkalinity.[15, 98] This approach was designed to be conservative, the goal being that very few homes in waters labeled "non-aggressive' would test above the 1.3 mg/L copper AL after a few months of exposure to the water. Ultimately, the most recent LCR revisions did not follow these NDWAC recommendations attempting to address cuprosolvency concerns. The framework can still provide guidance to building managers, residents, or utilities who wish to address these concerns on a voluntary basis.[27, 30]

This study aims to guide voluntary action addressing cuprosolvency concerns in new copper plumbing by identifying the minimum orthophosphate doses needed to maintain copper below 1.3 mg/L as a function of the water's alkalinity.[44, 116, 137, 304] This proposed NDWAC approach was vetted and refined using experiments with copper pipe. Simpler and less expensive laboratory cuprosolvency tests monitoring the aging of fresh cupric solids were studied as part of this evaluation.

3.2 Methods

Bench scale cuprosolvency testing using copper tube segments was used to establish minimum doses of orthophosphate to render a water "non-aggressive" to copper as a function of pH and alkalinity. "Non-aggressive" is defined as a water which would reduce soluble copper below the 1.3 mg/L AL after 5.5 months or less of exposure for a typical copper pipe.

3.2.1 Copper tube cuprosolvency testing

Copper tube cuprosolvency tests utilized 8.5" segments of new 1/2" diameter type M copper tubing. Each test utilized 5 pipe segments purchased from 4 different manufacturers (20 segments total). Pipes were stoppered at one end, filled with water, and covered during storage to prevent air exchange. Water changes were performed using a dump and fill protocol three times per week to simulate flow inside pipes, with periods of longer stagnation due to COVID-19 related laboratory shutdowns. Experiments were carried out using synthetic drinking waters under conditions believe "worst-case" (i.e. likely to elicit high cuprosolvency), including cooler temperature (15 °C) and the presence of trace NOM (0.2 mg/L NOM as fulvic acid from International Humic Substance Society) and high sulfate (200 mg/L).[44, 106, 111, 119, 155] Experiments utilized 4 waters with representative combinations of pH and alkalinity, with 4 orthophosphate doses per condition (16 total waters) (Table 3.1). A total of 320 tube segments were tested. Periodic composite (combined water from all tubes for a water condition) and full (individual tubes) samplings were performed, and samples analyzed for copper concentrations released into the water.

Table 3.1: **Summary of synthetic water conditions for cuprosolvency testing in copper tube.**

Alkalinity (mg/L as $CaCO_3$)	pH	Orthophosphate Dose (mg/L as P)
35	7.25	0
		0.06
		0.13
		0.26*
100	7.25	0
		0.2
		0.4
		0.6
250	7.5	0
		0.5
		1
		1.5
500	8	0
		1.5
		2
		2.5

* After Week 20 the orthophosphate dose was decreased to 0.03 mg/L as P

3.2.2 Cuprosolvency testing using fresh copper solids

An attempt was made to determine the minimum orthophosphate dose using freshly prepared cupric solids. A simplified cuprosolvency test was used based on Edwards et al. (2001),[106] but without the automatic titration method of prior testing,[206, 207] to facilitate easier application by utilities and other stakeholders. Initial experiments utilized analogous water chemistries as in copper tube experiments (Table 3.1) with 0.4 mg/L NOM. Cupric solid formation was initiated with the addition of 0.5 mM cupric nitrate while stirring at 200 rpm. The pH was adjusted to the target (+/- 0.05) within 5 minutes and maintained within +/- 0.25 of the target thereafter using 1 M NaOH or nitric acid. Aliquots were stored in plastic

bottles until sampling at predetermined intervals (n=1 for samples up to one week; n=5 replicates for 2-week and 1-month samplings). Bottles were shaken at each sampling time. Dissolved copper was determined as solids aged by filtering samples through 0.45 µm nylon syringe filters (Whatman) prior to analysis. Composite solids samples were collected after 2 weeks and 1 month of aging using a vacuum filter (Whatman 0.45 µm).

Additional tests examined orthophosphate dosing strategies. Experiments utilized 1L of test water at pH 7.5 with 250 mg/L as $CaCO_3$ alkalinity, 0.2 mg/L NOM, and 200 mg/L sulfate. First, a 1.5 mg/L as P orthophosphate residual was maintained by adding an initial dose of orthophosphate with additions at each sampling time to replenish orthophosphate to the target level. The second experiment employed one initial orthophosphate addition of 4.3 mg/L as P, equivalent to the total orthophosphate added in the previous test. Samples were filtered using a 0.45 µm pore size filter for dissolved copper analysis.

3.2.3 Analytical methods

Alkalinity and pH were determined via standard methods.[72] Hach methods 8506 and 8048 were used to track copper and orthophosphate concentrations (Hach DR3900). All samples used for statistical analysis were analyzed using Atomic Absorption Spectroscopy (AAS, Perkin-Elmer 5100 PC AAS) via method 3111 B (copper) and ICP-MS/MS (Thermo Scientific iCAP RQ ICP-MS) via method 3125 B (phosphorous, and other metals), after acidification to 2% nitric acid by volume for at least 16 hours.[72] QA/QC was performed for AAS and ICP-MS/MS after every 10 samples. All materials were reagent grade quality.

3.2.4 Data Analysis

Statistical analyses were performed on full sampling results from individual tube segments using R statistical software (version 4.2.1) and the ANOVA function with a significance threshold of $p \leq 0.05$. Interpolation of full sampling results facilitated development of linear regressions identifying minimum orthophosphate doses required to yield copper below the 1.3 mg/L AL for given alkalinities considering the highest, second highest, and third highest copper concentrations (i.e., 90^{th}, 95^{th} and 100^{th} percentile for 20 replicates). Copper solubility at thermodynamic equilibrium was predicted using MINEQL+ (version 4.6) following the method in Section B.2.

3.3 Results and Discussion

3.3.1 Proposed utility framework for cuprosolvency mitigation

A framework was developed, stemming from NDWAC consensus statement suggestions, to guide proactive utilities and other stakeholders in addressing cuprosolvency issues (Figure 3.1). The NDWAC proposed minimum orthophosphate doses for given alkalinities that would yield "non-aggressive" waters and almost always keep copper below the 1.3 mg/L AL even in relatively new homes.[15] Such "minimum" doses were intended to serve as criteria for effective cuprosolvency control without requiring burdensome sampling to verify performance. We refined that "minimum" orthophosphate dose criteria and used it as the basis for our framework.

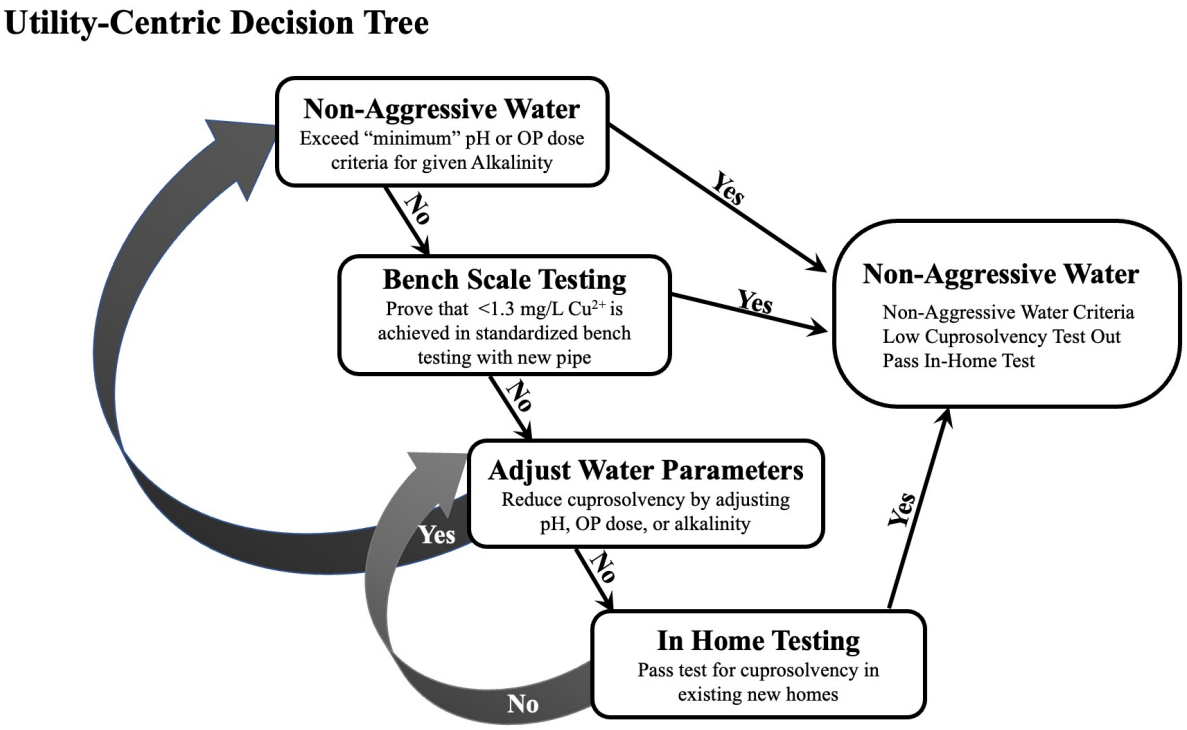


Figure 3.1: **A proposed decision-making framework for utilities to aid with determination of "non-aggressive" waters for new copper piping**

We posited that if a utility did not meet the "minimum" criteria, which was designed to have a safety factor, they could prove that their waters were non-aggressive using verification testing. Such testing could be done for their particular water using simple bench-scale tests with worst case new copper pipe, or through traditional field sampling, as prescribed under the LCR but sampling in homes with new copper pipe. In the absence of regulations these approaches might someday qualify as best practices identified in "Recommended Standards for Water Works" or similar programs.[360]

3.3.2 Cuprosolvency Testing

Effects of orthophosphate in copper tube cuprosolvency testing

Orthophosphate addition to the synthesized water was successful in yielding average copper concentrations below the 1.3 mg/L AL for all waters after the end of week 1, excepting one condition, which was only 2.4% above the AL (Figure 3.2). Consistent with the underlying NDWAC "minimum" orthophosphate criteria hypothesis, higher orthophosphate doses generally resulted in lower cuprosolvency. The exception was the lowest alkalinity, where elevated copper was not a problem; so no orthophosphate would be required.

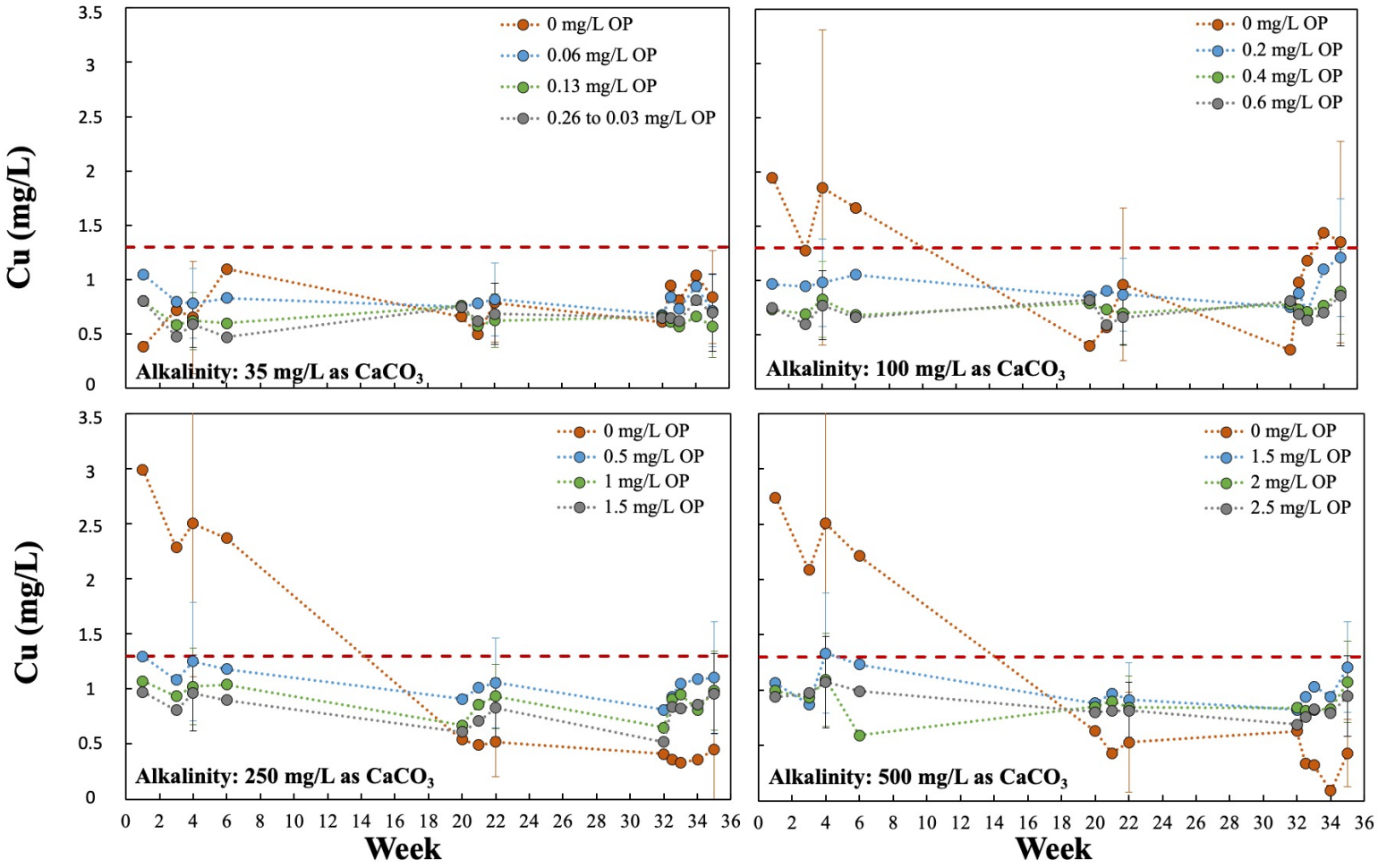


Figure 3.2: **Mean copper release from new copper pipes treated with varying levels of orthophosphate corrosion control.** (n=20 for weeks 4, 22 and 35; n=1 composite for all other times). Error bars represent one standard deviation. Red dotted lines denote the 1.3 mg/L action level. All orthophosphate (OP) doses are in mg/L as P.

Most orthophosphate treated waters yielded average copper below those without orthophosphate within the first six weeks of treatment, but as expected, in some waters orthophosphate yielded higher long-term cuprosolvency. For example, initial copper concentrations (first six weeks) at pH 7.5 and 250 mg/L as $CaCO_3$ alkalinity with orthophosphate were less than half that without orthophosphate (0.94 – 1.07 mg/L vs. 2.29 – 2.99 mg/L), but after 22 weeks the condition with orthophosphate had higher copper than without orthophosphate (0.93 vs. 0.52 mg/L). Such long-term detriments of orthophosphate dosing have been noted in prior literature, and are attributed to rapid formation of low solubility cupric phosphates, but inhibited formation of very low solubility minerals like tenorite or malachite that may form over a period of months, years or decades.[44, 106, 114, 116, 137, 155, 179, 185, 204, 299, 301, 303, 304, 339] Because of potential higher long-term copper release and concerns about expense and sustainability of phosphate use, utilities could instead choose to prove the efficacy of pH/alkalinity corrosion control criteria in their water.[44]

Visual observations of scales were consistent with these results, with blue-green scales characteristic of malachite present in high alkalinity pipes without orthophosphate whereas no blue-green scales were observed in pipes conditioned with orthophosphate.[44]

Manufacturer differences in copper tube cuprosolvency testing

Contrary to expectations that most off-the-shelf copper tube would yield consistent and relatively similar cuprosolvency, substantial variability was observed amongst tube of different manufacturers. Mean copper concentrations from one manufacturer were 0.39 to 1.26 mg/L (average 69%, range 51-88%) lower than those of the other three manufacturers and were significantly different ($p < 0.05$, n=5 per manufacturer) for all OP conditions and times tested. In contrast, mean copper concentrations comparing the other manufacturers for these conditions were only significantly different for 17.6% of cases, with an average difference of 10%

(range 0-46%). These cuprosolvency differences may result from manufacturing and atmospheric differences creating different oxide films initially present on the pipes.[184] Using tube from the outlier manufacturer in cuprosolvency testing could be misleading, since it yielded significantly lower copper (<AL) for 19.4% of cases where the mean copper from other manufacturers exceeded the AL. This complication did not affect determination of "minimum" orthophosphate criteria using the 20 tubes from all four manufacturers presented in the next section.

Determination of minimum orthophosphate dose criteria

The minimum orthophosphate doses that yielded copper below the 1.3 mg/L AL were determined for each alkalinity (Figure 3.1, example method in Section B.1). To be conservative, the highest, second highest, and third highest copper concentrations (100^{th}, 95^{th} and 90^{th} percentiles) of the 20 pipe segments were utilized for determining orthophosphate criteria. Data after four and 22 weeks of exposure for three orthophosphate doses were interpolated to determine the orthophosphate doses needed at each alkalinity to achieve copper below the 1.3 mg/L AL. "Worst case" linear regressions (Figure 3.3) were developed from interpolated results using the highest value after either 4 weeks or 22 weeks of pipe aging. Regressions demonstrated good correlations ($R^2 > 0.98$) and generally good agreement between 90^{th}, 95^{th} and 100^{th} percentile regressions. Results verify that increasing orthophosphate doses are required to reduce cuprosolvency at higher alkalinities as predicted by Schock et al.[302, 304]

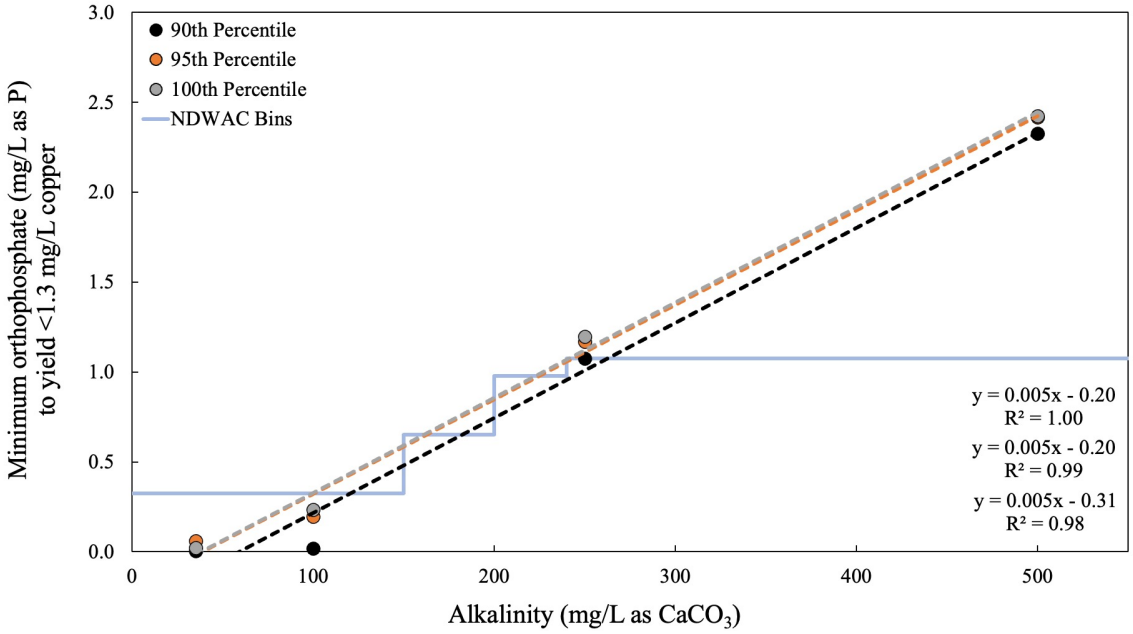


Figure 3.3: **Linear regressions defining the "minimum" orthophosphate criteria for varying alkalinities to define waters "non-aggressive" to copper.** Orthophosphate values represent the highest interpolated value, after either 4 or 22 weeks of testing, from linear correlations of orthophosphate and maximum (100^{th} percentile), second (95^{th} percentile) and third (90^{th} percentile) highest copper concentration for each alkalinity tested.
The bin system recommended by the NDWAC consensus statement is noted by the light blue line.[15]

These regressions provide a conservative and continuous method to help utilities estimate required orthophosphate doses needed to control cuprosolvency as a function of alkalinity, under what are predicted to be relatively worst-case water chemistry conditions in terms of sulfate and NOM. Results also demonstrate relatively good agreement with the specific minimum orthophosphate levels tentatively recommended by the NDAWC consensus statement (Figure 3.3).[15] However, for the water chemistries tested, the NDWAC bins may not be conservative enough to control cuprosolvency for higher alkalinity waters within each bin. This highlights the value of the continuous regression approach for more precise estimation of "minimum" orthophosphate doses needed based on a water's maximum alkalinity to avoid

situations with sustained high cuprosolvency.

Our results suggest a linear correlation ($R^2 > 0.98$) between alkalinity and the minimum orthophosphate dose within the range of water conditions tested. Modeling of copper solubility at equilibrium with $Cu_3(PO_4)_2$ solid using Mineql+ also indicated a quasi-linear relationship at a given pH, with model predictions deviating from linear regressions by 16% or less (pH 7.25 and 8, and alkalinities >100 to 500 mg/L as $CaCO_3$) (Figure B.3). However, when considering Mineql+ model predictions corresponding to water conditions used to develop "minimum" orthophosphate criteria, the Mineql+ model had a slope over 4 times higher than that of the "minimum" pH linear regressions developed by experiment herein (Figure B.2). Other literature also notes inaccuracies in predicting soluble copper based on orthophosphate dose from chemical equilibrium modeling, so Lytle et al. developed an empirical model, which fit data from a fresh-solids cuprosolvency method.[206] That model predicts much lower orthophosphate doses than that identified from our "minimum" orthophosphate criteria developed from pipe testing.

3.3.3 Development of a simplified fresh solid cuprosolvency test method

A cuprosolvency test method was developed using fresh copper solids. Although cuprosolvency tests using fresh solids may pose advantages, there were several complications that precluded their use for developing analogous minimum orthophosphate dose criteria as developed using copper tube in the previous section. Specifically, in initial tests, when only a single dose was used, the orthophosphate was consumed and the residual that controls solubility varied. Two modified methods were used to maintain higher phosphate residuals during the test.

Advantages of cuprosolvency testing using fresh copper solids

Simplified bench cuprosolvency testing, that starts with fresh cupric solids may pose several advantages over testing using copper tube. In new pipes, cupric hydroxide solids are thought to form from cuprous solids over a period of days, weeks, or months, so it could take time for such solids to accumulate and control copper dissolution.[106, 155] Once they form, these cupric hydroxides can then begin the aging process. Starting the testing with fresh $Cu(OH)_2$ particles, would possibly accelerate that process, allowing solids aging to be observed more quickly, with 6.7 times less effort, 30% less initial cost, and up to 7 times lower testing cost than in copper tube tests (Table 3.2, Section B.3).[106, 155] Although in-home testing may be the least costly approach for utilities to detect cuprosolvency problems on a per home basis, it may require testing of many homes and coordination with homeowners to demonstrate that cuprosolvency is not a problem, in which case an alternative approach using the fresh solids method would be of value.

Table 3.2: **Cost estimates detailed in Section B.3,** are on per-home basis or for initial experiment. Costs for subsequent tests account for initial materials and extra consumables that could be used in additional experiments. *Assumes access to ICP-MS/MS ($6 per sample), pH meter, and wages of $20 per hour.

	Materials				Time			Overall		
Apparatus	**Initial Materials**	**Consumables**	**Initial Materials Cost**	**Consumable Cost**	**Required Tasks**	**Labor**	**Labor Costs**	**Duration**	**Cost For Initial Test**	**Cost For Subsequent Tests**
Fresh Solids Test	Carboy, bottles, Stir plate	Reagents, syringes, filters, analysis	$1,100	$1,600	Preparing water and particles, sampling, sample analysis	12 hr.	$240	30 days	$2,900	$370
Copper Tube Test	Carboy, Temperature Control, Weighted lid, tray, bottles, pipe reamer	Reagents, stoppers, pipe segments, rubber bands labels, plastic wrap, cups, analysis	$1,400	$1,100	Preparing pipes, water changes, sampling, sample analysis	80 hr.	$1,600	6 months	$4,200	$2,600
In-Home Test	Bottles	Reagents, analysis	$80	$200	On-site sampling, sample analysis	6 hr./visit	$120	1 week	$400	$180

Initial testing using fresh copper solids

Initial cuprosolvency tests using fresh copper solids were tested using slight modifications to a prior simple method.[106] Results indicated that aging and scale formation may progress faster than in tube-based tests. In tests with fresh solids, copper concentrations fell well below the AL (0.09 to 0.50 mg/L dissolved copper) within 1 month (Figure 3.4) for all alkalinity conditions without orthophosphate, but were consistently higher in tube experiments even after 35 weeks of aging (0.65 to 2.50 mg/L after 1 month, 0.43 to 1.35 mg/L after 35 weeks). This indicates that particle-based methods may not be indicative of short-term cuprosolvency behavior in new pipe which can cause acute health concerns, but may be more representative of the final mineral product achieved with long term aging. It is unclear how long it would take to achieve these longer-term results in copper tube testing with regular water changes.

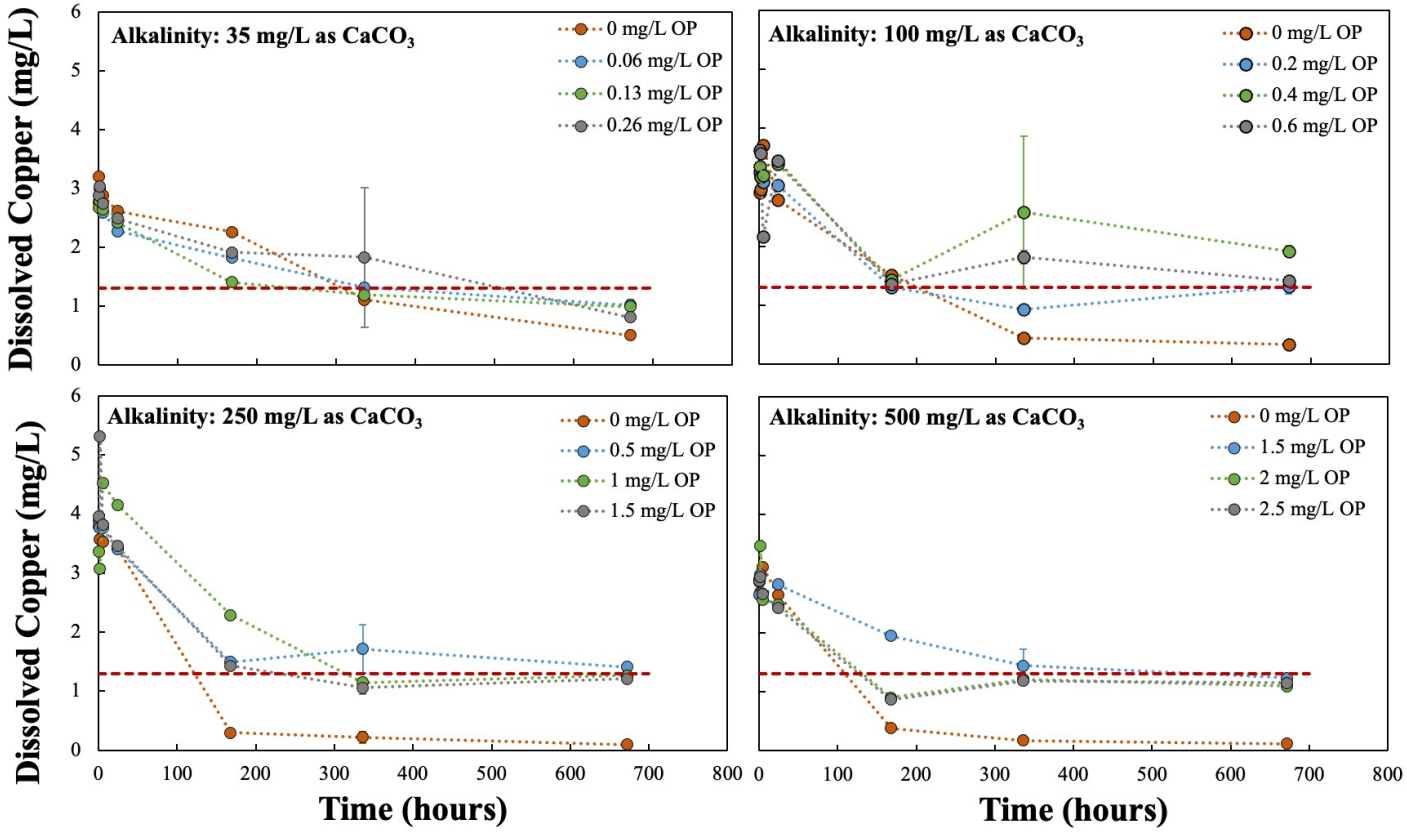


Figure 3.4: **Mean dissolved copper release from copper particles treated with varying levels of orthophosphate corrosion control.** (n=5 for weeks 2 and 4; n=1 for all other times). Red dotted lines denote the 1.3 mg/L action level. All orthophosphate (OP) doses are in mg/L as P.

One challenge of applying short-term simple cuprosolvency tests in the presence of phosphate inhibitors, is simulating orthophosphate residuals observed in copper pipes during stagnation in real systems. Specifically, in new pipes orthophosphate is depleted from the water as it is incorporated into pipe scale, whereas upon further flow and scale accumulation orthophosphate is not significantly depleted during stagnation. For instance, in copper tube experiments with orthophosphate replenishment via water changes, 16-32% of orthophosphate was taken up in 100 mg/L as $CaCO_3$ alkalinity waters at the end of the first week of testing, but only up to 11% was taken up after 3 weeks of testing. In fresh solids tests without orthophosphate replenishment, 65-98% of the orthophosphate was consumed by the end of the 1-month experiment.

Thus, in the initial fresh solids protocol without replenishing orthophosphate, cuprosolvency was an average of 0.42 mg/L higher (difference 0.12 to 1.22 mg/L, range 0.81 to 1.92 mg/L) than in corresponding copper tube tests (22 weeks, range 0.62 to 1.06 mg/L). It was nonetheless considered promising even when underdosing phosphate that, analogous to copper tube testing, orthophosphate had little effect on cuprosolvency at low alkalinity and yielded higher long-term cuprosolvency than conditions without orthophosphate at high alkalinity. These results are consistent with the short-term benefit and long-term detriment resulting from the formation of higher solubility cupric phosphate scales.[44, 106, 114, 116, 137, 155, 179, 185, 204, 299, 301, 303, 304, 339]

3.3.4 Modified cuprosolvency test using fresh copper solids

The cuprosolvency method using fresh copper solids was modified to overcome the orthophosphate demand of the aging solids. The modified method utilized one large batch of test water (pH 7.5, 250 mg/L as $CaCO_3$ alkalinity, 0.2 mg/L NOM, and 200 mg/L sulfate) and either

one large dose of orthophosphate, or supplemental orthophosphate added throughout the test. These dosing strategies yielded variable cuprosolvency results in comparison to previous cuprosolvency test methods.

When successive dosing was performed, orthophosphate was added to maintain a concentration of about 1.5 mg/L as P. Results demonstrate initial depletion of orthophosphate coupled with reductions in soluble copper (Figure 3.5), as may be expected if cupric phosphate solids were rapidly forming and controlling cuprosolvency.[106, 304] Orthophosphate and copper concentrations remained relatively stable after 2 hours of reaction time, confirming that a steady state condition was achieved much faster than in the tube tests that first require a reservoir of cupric solids to form. The final dissolved copper concentration of 0.53 mg/L (17 hours) was just over half that in the copper tube-based test (0.96 mg/L after 35 weeks) with similar conditions and residual orthophosphate concentration. The additional orthophosphate and potential faster kinetics in this test may have facilitated more complete formation of a cupric phosphate scale than in previous tube or fresh solids tests.

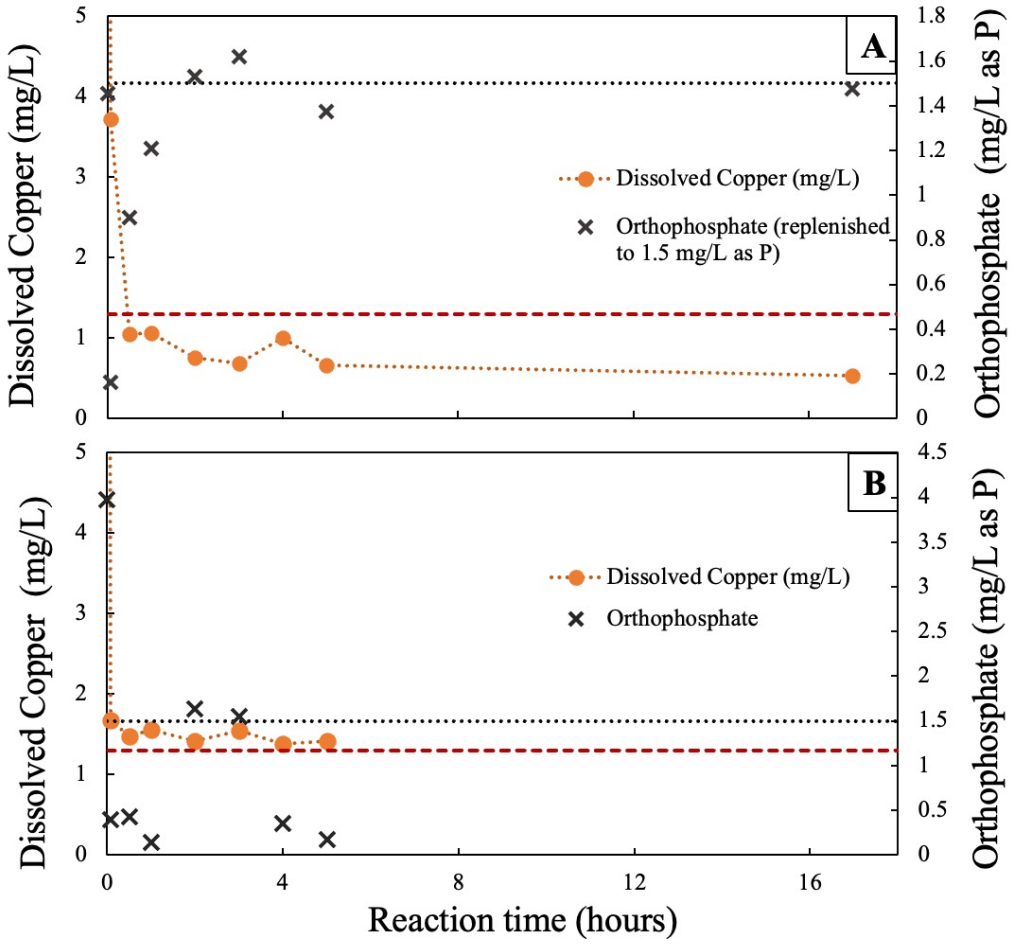


Figure 3.5: **Dissolved copper release from copper particles treated with 4.3 mg/L as P orthophosphate corrosion control.** A. Added to replenish orthophosphate concentration to 1.5 mg/L as P and B. Added initially all at once. Initial copper concentrations are 0.5mM or 31.8 mg/L.
Red dotted lines denote the 1.3 mg/L action level. Black dotted lines denote the target residual orthophosphate concentration. All orthophosphate (OP) doses are in mg/L as P.

In contrast, when the same total orthophosphate (4.3 mg/L as P) was added as one initial dose, residual orthophosphate quickly fell below the 1.5 mg/L as P target, yielding almost three times higher copper (1.41 mg/L dissolved copper, 5 hours) than when the target residual was maintained at a constant level during the experiment. Even though orthophosphate could have been limiting in both tests, with less than half the orthophosphate added that is

stoichiometrically needed to form cupric phosphate, the higher residual orthophosphate and lower cuprosolvency in the successive dosing test could result from formation of larger particles, reducing the available surface area for reaction with orthophosphate. Overall, these results demonstrate that the timing and dose of orthophosphate addition can affect particle formation via orthophosphate uptake and cuprosolvency. Further, they highlight the need for developing a standardized particle cuprosolvency test method that accounts for factors like solution mixing and maintaining a target residual.

3.4 Implications

Although the gaps in copper corrosion control were not addressed in the recent LCR revisions,[27, 30] proactive utilities may choose to address sustained elevated cuprosolvency to protect public health or alleviate consumer complaints. For example, it may be relatively easy for proactive utilities to add copper tube testing, to standard CCT optimization studies with lead pipe loops or bench testing.[177] This work expands upon the NDWAC recommended guidance to help utilities predict the orthophosphate doses needed to address sustained elevated cuprosolvency in their distribution systems. Utility use of this framework, as well as the development of faster and simpler, fresh solid cuprosolvency test methods, could help to quickly identify water conditions likely to yield "low cuprosolvency" water without costly in-home testing, potentially saving utilities time and resources. Although results indicate that orthophosphate may not be necessary to control cuprosolvency in the short-term in lower alkalinity waters, or even in certain higher alkalinity waters that age to lower solubility solids, regressions were developed to help determine orthophosphate doses needed for circumstances where cuprosolvency is problematic. One advantage of this "minimum" orthophosphate approach is that the systems reached a pseudo steady state relatively quickly, indicating that

utilities can expect to see a characteristic response within weeks of orthophosphate addition.

It is also important to note that orthophosphate addition for corrosion control can have multiple benefits. First, orthophosphate is often added to control corrosion of lead and iron, in which case its effect on copper is a side benefit if it reduces cuprosolvency, or a detriment if it increases cuprosolvency.[44, 116, 137, 304] Orthophosphate addition can be expected to be effective even in higher alkalinity waters without the potential for causing precipitation of carbonate scales that would arise from raising the pH.[137] In fact, in those situations, orthophosphate can reduce the scaling potential of the water.[209]

Finally, orthophosphate treatment yields nearly immediate reductions in cuprosolvency, with low and relatively stable copper concentrations possible after less than 1 week of treatment, which could be particularly beneficial for utilities experiencing abrupt changes in water source or quality. However, the use of orthophosphates also has certain drawbacks. For example, continuous orthophosphate addition is required to maintain cuprosolvency reductions, with higher concentrations sometimes needed to control cuprosolvency than to control lead release.[44, 302] These high orthophosphate doses could make it difficult for utilities to effectively control cuprosolvency while meeting wastewater phosphate discharge limits and pose concerns about expense and sustainability of phosphate use.[44, 303] Further, the effectiveness of orthophosphate treatment and dose needed is expected to vary with water quality, including pH, alkalinity, and other parameters.[98, 299, 302, 304] As observed in this study, higher orthophosphate doses are required for higher alkalinity to achieve cuprosolvency reductions. Finally, this study confirms that some cases, despite the initial benefit, orthophosphate use can lead to higher long term cuprosolvency than conditions where pipes age normally and form lower solubility solids.[44, 106, 114, 116, 137, 155, 179, 185, 204, 299, 301, 303, 304, 339]

More research could improve the practice of optimizing orthophosphate doses. For example, further mechanistic study, investigation of thermodynamic data, and research on spectroscopic phase identification for X-ray amorphous solids for various orthophosphate minerals would aid in understanding and modeling orthophosphate effectiveness and doses needed.[304] Very high levels of NOM have been reported to reduce the efficiency of orthophosphate corrosion inhibitors, indicating that higher orthophosphate doses may be needed in some situations for cuprosolvency control.[44, 193] Further, the effectiveness of blended phosphates or polyphosphates for cuprosolvency control warrants further investigation, as they have been suggested to complex copper and were associated with sustained elevated copper above the AL, even while the utility was in compliance with the LCR.[58, 302] An improved cuprosolvency test using fresh copper solids could be helpful for more quickly evaluating the effects of these parameters, however it requires further development to overcome challenges maintaining constant residual concentrations to mimic the replenishment of constituents as occurs in pipe systems with flowing water. Finally, it would also be beneficial to determine the "minimum" pH vs. alkalinity regression criteria, suggested by the NDWAC, which would allow utilities to simultaneously consider either orthophosphate or pH/alkalinity control methods.

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Chapter 4

Challenges Predicting Cuprosolvency in Drinking Water Based on Common Water Chemistry Parameters

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Abstract

Elevated cuprosolvency in drinking water can pose aesthetic and health concerns for residents and their pets. Gaps in the Unites States Environmental Protection Agency Lead and Copper rule leave some consumers vulnerable to high cuprosolvency water for the foreseeable future, creating a need to guide stakeholders who wish to address this concern. We develop linear regressions that can help identify a "minimum pH" target for given alkalinities to achieve "acceptable cuprosolvency" for most waters. But we also identify confounding effects from calcite precipitation and demonstrate that NOM, silica, and sulfate concentrations unpredictably affect pipe aging. Some waters may require use of orthophosphate to control corrosion or other interventions to avoid problems with elevated cuprosolvency.

4.1 Introduction

Elevated cuprosolvency in drinking water resulting from corrosion of copper plumbing can create aesthetic issues and health concerns for consumers and their pets.[8, 115, 304, 329] The United States Environmental Protection Agency (USEPA) set an action level (AL) of 1.3 mg/L for 90^{th} percentile copper concentrations in the Lead and Copper Rule (LCR) to address systemwide issues with elevated cuprosolvency.[8, 44, 49, 339] However, gaps in the LCR leave many residents at risk, especially in non-residential buildings, private well water, and in homes with new copper pipes which tend to have higher cuprosolvency.[44, 49, 99, 115, 270, 304, 339] The National Drinking Water Advisory Council (NDWAC) consensus statement attempted to close some of these gaps in proposed LCR updates. Specifically, they suggested that a "minimum" pH or orthophosphate dose for a given alkalinity could classify waters as "non-aggressive" to new pipe (Table 4.1), thereby allowing effective cuprosolvency control to be achieved without any utility requirement for verification sampling in homes.[15]

Table 4.1: **Suggested NDWAC minimum pH criteria for corrosion control based on the level of alkalinity in the distributed water.**[15]

Suggested NDWAC pH Criteria	
Alkalinity (mg/L as $CaCO_3$)	pH
< 35	> 7.0
36 - 100	> 7.2
101 - 150	> 7.5
151 - 250	> 8.0

The scientific basis for the NDWAC minimum pH approach is based on the premise that parameters such as pH and alkalinity exert primary control on cuprosolvency in the absence of phosphate. Copper surfaces in pipes initially react with water to form cupric hydroxide [$Cu(OH)_2$] and other solids that can have high cuprosolvency. With time these solids can transition to lower cuprosolvency minerals including malachite.[44, 114, 185, 304] Copper solubility is almost always predicted to decrease with increasing pH because the pH affects corrosion rate, hydrolysis reactions, the identity of cupric solids that are thermodynamically favored, and the rate of transitions to less soluble phases (i.e., scale aging).[44, 98, 114, 155, 304, 347] In the short-term higher alkalinity almost always increases cuprosolvency via formation of soluble cupric complexes with bicarbonate, but in the longer-term increases in alkalinity can reduce cuprosolvency by contributing to the formation of relatively insoluble

malachite scales.[98, 114, 302, 304, 347] These time dependent, aging effects, of pH and alkalinity are complex.[155, 302, 304] Some chemical models, empirical models, and field data nonetheless indicate that sometimes there is a linear relationship between higher alkalinity and higher cuprosolvency in new and old copper tube.[114, 346] To address these problems it was proposed that the "minimum pH" would increase with higher alkalinity.

Many other water quality parameters have been reported to affect cuprosolvency. Parameters such as free chlorine, chloride, sulfate, hardness, temperature, natural organic matter (NOM), microorganisms and nitrogen, sulfur, and iron species can influence cuprosolvency through a variety of mechanisms.[113, 155, 302, 304, 347] Even though the literature predicts that relatively common sulfate minerals should eventually transition to more stable tenorite and malachite scales, empirical data in one study showed that sulfate had a significant linear relationship with cuprosolvency in aged copper pipes.[185, 346, 347] This increased cuprosolvency in the presence of sulfate was also demonstrated in one laboratory study.[106] Silica was also reported to affect cuprosolvency via formation of silicate minerals and effects on scale structure.[264] Further, NOM was reported to have divergent effects, increasing cuprosolvency via complexation, mobilization of colloids, and interfering with passivating scale formation, but sometimes reducing cuprosolvency via adsorption, and stabilizing copper(I).[44, 111, 113, 179, 304] The simple NDWAC approach did not account for these "secondary" factors that influence cuprosolvency.[98]

This study sought to help proactive utilities address their customers' elevated cuprosolvency issues by investigating the "minimum" pH needed to maintain copper below the 1.3 mg/L AL in water of varying alkalinity. Monitoring results from utilities collected over the last three decades were inspected to verify our conceptualization of these problems. We then identify significant limitations to this approach and evaluate other aging and water quality factors that can be influential. Finally, to assist stakeholders including consumers, utilities

and regulators attempting to deal with these problems, we propose updated guidance that attempts to account for some of these uncertainties.

4.2 Methods

Bench scale cuprosolvency tests using copper tube were used to establish the minimum pH needed to result in a water that is "non-aggressive" to new copper as a function of alkalinity. We defined "non-aggressive" waters as those resulting in copper below the 1.3 mg/L action level after 22-35 weeks of pipe aging. Further, we used copper tube and fresh copper solids to examine the effects of other water quality parameters on cuprosolvency.

4.2.1 Copper tube cuprosolvency testing

Copper tube cuprosolvency tests followed the same experimental protocol used in Kriss et al.(2023).[181] Tests utilized 8.5" segments of new ½" type M copper tube filled with synthetic drinking waters, and followed similar water change, storage protocols and "worst-case" background water conditions.[44, 106, 111, 119, 155]

Copper tube tests were used to establish the "minimum" pH needed to yield waters "non-aggressive" to new copper. Experiments utilized a synthetic drinking water with 200 mg/L sulfate and 0.2 mg/L fulvic acid from the International Humic Substance Society at 4 alkalinities. Each water was adjusted to 3 pH values (12 water conditions summarized in Table 4.2). The lowest pH for three of the four alkalinities was increased in Phase 2 in an attempt to achieve cuprosolvency below the AL in at least one water condition for each alkalinity. Waters were tested in 5 pipe segments from each of 4 manufacturers (n=20 per water condition, 240 total segments). Water changes followed a regular three time per week schedule

with some periods of longer stagnation due to laboratory shutdowns related to COVID-19. Periodic full and composite sampling was performed.

Table 4.2: **Summary of synthetic water conditions for establishing "minimum" pH criteria**
*The lowest pH values for each of the three highest alkalinities were increased for Phase 2 (blue values).

Experimental Conditions		
	Phases 1: Weeks 0-20	Phase 2: Week 20-35
Alkalinity (mg/L as $CaCO_3$)	pH	pH
35	7 7.25 7.5	7 7.25 7.5
100	7 7.25 7.5	**7.8** 7.25 7.5
250	7.25 7.5 7.75	**8.25** 7.5 7.75
500	7.75 8 8.35	**8.6** 8 8.25

Additional copper tube tests investigated the effects of NOM and sulfate on cuprosolvency. Tubes from one manufacturer (n=5) were used to test the cuprosolvency of 14 simulated drinking waters, consisting of seven NOM concentrations (0 to 2 mg/L as fulvic acid from the International Humic Substance Society) at two pH values (7.5 and 8.1). The higher pH was predicted to be above the "minimum" pH criteria and the other was below it. Experiments utilized water with an alkalinity of 250 mg/L as $CaCO_3$ and initially had 200 mg/L of sulfate. After 33 weeks, sulfate levels were decreased to 0 mg/L for the 3 highest cuprosolvency tubes at each condition. Selected samples were filtered using a 0.22 µm syringe

filter to differentiate particulate and dissolved copper.

4.2.2 Cuprosolvency testing using fresh copper solids

Cuprosolvency tests using freshly prepared cupric solids were performed using methods similar to prior research.[106, 181] Experiments were initiated with the addition of 0.5 mM cupric nitrate in 1L bottles with synthetic drinking water consisting of 1 mM $NaNO_3$ and varying pH, alkalinity, sulfate, silica, and NOM (fulvic acid from IHSS) concentrations (Table 4.2). Silica and/or sulfate were added just after cupric nitrate addition and within 5 minutes the pH was re-adjusted to the target (+/- 0.05) and maintained (+/- 0.25) for the duration of the experiment using sodium hydroxide or nitric acid. Bottles were stirred at 200 rpm for 1 hour during experiment initiation and during subsequent sampling and were mixed using an end over end mixer for 2 weeks then transferred to a shaker table at 100 rpm for the duration of the trial. Samples were collected at predetermined times and passed through 0.45 µm pore size nylon syringe filters (Whatman) to determine dissolved copper concentrations. At the end of each trial, solids were collected for XRD analysis via filtering remaining water with a vacuum filter (Whatman 0.45 µm filter) and rinsing 3 times with 100 mL of DI water.

4.2.3 Analytical methods

Alkalinity and pH were determined via standard methods.[72] Copper was determined using Atomic Absorption Spectroscopy (AAS, Perkin-Elmer 5100 PC AAS) via method 3111 B and using ICP-MS/MS (Thermo Scientific iCAP RQ ICP-MS) via method 3125 B.[72] Samples were acidified to 2% nitric acid by volume for at least 16 hours prior to analysis and included QA/QC after every 10 samples. All materials used were of reagent grade quality.

4.2.4 Data analysis

Development of the "minimum pH" criteria is detailed in Section C.1 and followed analogous interpolation and regression methods similar to those used to determine the "minimum" orthophosphate criteria in Kriss et. al. (2023).[181] Statistical analysis was performed on full sampling results from copper tube testing using the ANOVA function (significance threshold $p \leq 0.05$) in R statistical software (version 4.2.1). MINEQL+ software (version 4.6) was used to model copper solubility at thermodynamic equilibrium.

4.3 Results and Discussion

The NDWAC consensus statement proposed that water quality criteria, based on a combination of either "minimum" orthophosphate or "minimum" pH, could be used to classify waters with effective corrosion control that were likely to result in copper below the 1.3 mg/L (or 1,300 ppb) EPA action level. Our prior study demonstrated that this approach may generally be effective for the phosphate criteria.[181] This work examines the effectiveness of the "minimum" pH approach.

4.3.1 Determination of "minimum" pH to yield "non-aggressive" water

Cuprosolvency in initial copper tube experiments with a synthesized water generally yielded lower cuprosolvency with increasing pH and longer aging time (Figure C.1). After one month of pipe aging, declining 90^{th} percentile cuprosolvency (i.e. the third highest copper value from n=20 pipe segments) was observed with increasing pH with one exception. This trend

held for lower alkalinity waters (35-100 mg/L as $CaCO_3$) through the end of the study. Statistically significant decreases in copper were also observed in 60% of cases from 1 to 8.75 months of pipe aging. These results are generally consistent with the underlying hypothesis for the "minimum" pH expectation, in that higher pH and longer pipe aging would generally yield lower cuprosolvency. However, a few cases did not follow expected trends. For example, 90^{th} percentile cuprosolvency was 3-4 times higher at pH 7.75 than at pH 7.5 in the 250 mg/L as $CaCO_3$ alkalinity water for months 5.5 and 8.75. Moreover, 14% of cases had increased dissolved copper from months 1 to 8.75.

The minimum pH values that yielded copper below the 1.3 mg/L AL were determined for each alkalinity (Figure 4.1A, Section C.1). Values were determined using either the 90^{th} 95^{th} or 100^{th} percentile copper (i.e., first, second, and third highest concentrations from n=20 tube segments) to allow for policy considerations such as use of a 90^{th} percentile AL under the current LCR. Following the same method of Kriss et al. (2023), data after 4, 22, and 35 weeks of aging for 3 pH values at each alkalinity were interpolated to determine the minimum pH needed to yield copper below the AL.[181] The highest interpolated values for each alkalinity were then used to develop "worst case" linear regression (Figure 4.1A), which exhibited good correlations ($R^2 > 0.98$) and generally good agreement between 90^{th}, 95^{th} and 100^{th} percentile results.

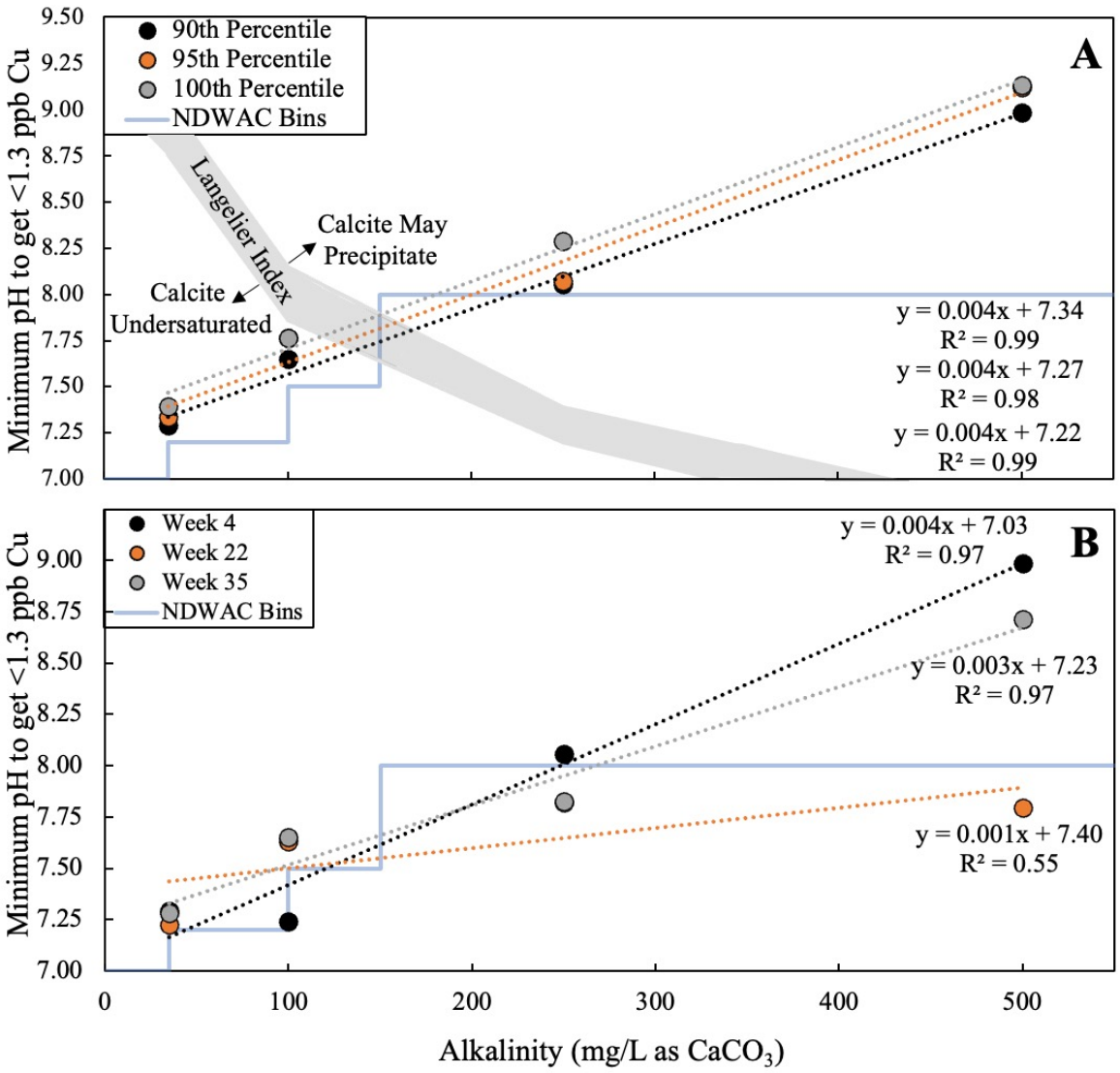


Figure 4.1: **"Minimum" pH needed to yield copper below the 1.3 mg/L action level for** A: Worst case values from the 90th, 95th, and 100th percentile copper using tubes and B: 90th percentile tubes after 4, 22, and 35 weeks of aging.
Values determined via pipe cuprosolvency tests (n=20 pipe segments per water condition). The Langelier Index was estimated via two methods in the AWWA WaterMath Corrosivity Estimator and one method from the Government of Newfoundland and Labrador.[24, 35]

The regressions confirm expectations that higher pH values will be needed to reduce cuprosolvency at higher alkalinities.[44, 98, 114, 155, 304] Moreover, results suggest that higher pH values may be needed to control cuprosolvency than initially predicted in the NDWAC consensus statement when using the "worst case" levels of sulfate and NOM tested for new copper tube in this work.[15]

4.3.2 Effectiveness of Corrosion Control Treatment in Long-term Utility Monitoring

We examined utility monitoring data since implementation of the LCR in 1991, to obtain insight into the effectiveness of CCT and aging processes. Our analysis of a 1992 AWWA survey identified only 20 of 361 utilities with 90th percentile copper concentrations exceeding the copper action level of 1.3 mg/L. Of the 20, 18 were not implementing corrosion control at the time.[98] Eight of these utilities eventually implemented pH adjustment as corrosion control. The other utilities implemented a combination of ortho/polyphosphate addition (n=3), an unspecified CCT strategy (n=3), or did not make significant changes for CCT (n=4). All of these utilities with available recent LCR monitoring data (n=17) reported 90th percentile copper concentrations below the action level after 2019 regardless of the CCT strategy implemented (Figure C.4). Eleven of the 16 had 90th percentile copper concentrations drop by more than 82% and below 500 ppb over the 27-30 intervening years. The five systems still reporting relatively high cuprosolvency after 2019 (560 – 1,270 ppb) included two utilities using pH-alkalinity CCT, one utility adding polyphosphates, two utilities with unspecified CCT, and one utility with no CCT. Overall, it is unclear whether CCT or pipe aging was responsible for the observed cuprosolvency changes over the last 3 decades.

The utility data generally corroborate the inverse relationship between pH and cuprosolvency, with lower 90th percentile copper concentrations observed at higher pH in the original survey.[98] Moreover, these utility results confirm, that the "minimum" pH criteria developed herein may be more conservative than needed in some cases. For example, three utilities were below the "minimum" pH criteria, but still had copper below the AL within 5 to 18 years without adding corrosion control treatment. Overall, these results are promising, but it is also unclear how consistently the reported utility pH values met the "minimum" criteria due to variations in water quality over time or the use of multiple sources.

This data also indicates that what constitutes a "reasonable" time frame for pipe aging to reduce copper to below the action level will be in the eye of the beholder. It is likely that some new residents will not be satisfied having to wait a few weeks, whereas other communities might want to wait decades if it avoids additional treatment costs. Herein, we operationally define a "reasonable" time frame as up to 35 weeks of pipe aging.

4.3.3 Challenges broadly applying "minimum" pH criteria

Our analysis revealed a variety of factors that could complicate broad application of the "minimum" pH criteria. These include a range of water quality considerations and some exceptional cases where cuprosolvency did not follow expected trends.

Potential for calcite precipitation

Before raising pH to control cuprosolvency, the potential for calcite precipitation and scaling should be considered, especially in high hardness and high alkalinity waters.[18] An illustrative boundary condition of the Langelier Index (LI) (i.e. LI=0) was estimated using three methods assuming all alkalinity is present in the form of $CaCO_3$ hardness (Figure

4.1A).[24, 35] Water with a pH exceeding the LI boundary condition would be oversaturated at 25°C and highly oversaturated at higher temperatures in water heaters, indicating that problems with scaling might occur.

Practically, calcite precipitation could be a concern for many surface waters that wish to control cuprosolvency. For instance, in one survey about 70% of surface waters had alkalinities greater than 100 mg/L as $CaCO_3$. Under our assumptions, alkalinities above this range, would likely have "minimum" pH criteria higher than the LI.[321] In other words, the threat of calcite precipitation might prevent use of a minimum pH approach in many high hardness and high alkalinity waters, because achieving that pH for corrosion control might cause excessive scaling. Utilities in that situation could instead consider using orthophosphate CCT and the previously developed "minimum" orthophosphate dose criteria.[181]

Variable cuprosolvency from different tube manufacturers

Tube manufacturer significantly affected cuprosolvency, consistent with manufacturing differences reported in the literature, which could have important implications for in-home and utility cuprosolvency tests.[184] Cuprosolvency results from Brand A were significantly lower ($p < 0.05$) from that of other brands for 74.5% of cases, while the other 3 brands were only different from each other 25% of the time. Brand A resulted in low cuprosolvency initially (average copper < 500 ppb for all waters at month 1), but in 5 of 12 cases there were statistically significant increases in copper of 300 ppb or more between months 1 and 8.75. The other 3 brands had higher initial copper for all water conditions, which generally decreased over time. Statistically significant increases in average copper with aging were only observed in 5 of 36 cases across the other 3 brands. These differences in observed cuprosolvency are exemplified in Figure 4.2, where cuprosolvency from Brand A increases over time but cuprosolvency from the other brands decreases over time. It is hypothesized that the dif-

ferent brands would eventually converge at some point, but if short-term testing had been done with Brand A tube alone, false conclusions about the effectiveness of pH would have been drawn. For instance, all samples from Brand A tube at 250 mg/L as $CaCO_3$ alkalinity and regardless of pH, yielded cuprosolvency less than the AL (<713 ppb) after one month of aging, which would indicate effective CCT, whereas cuprosolvency from the three other manufacturers was above the AL (1,800-4,650 ppb) for these conditions.

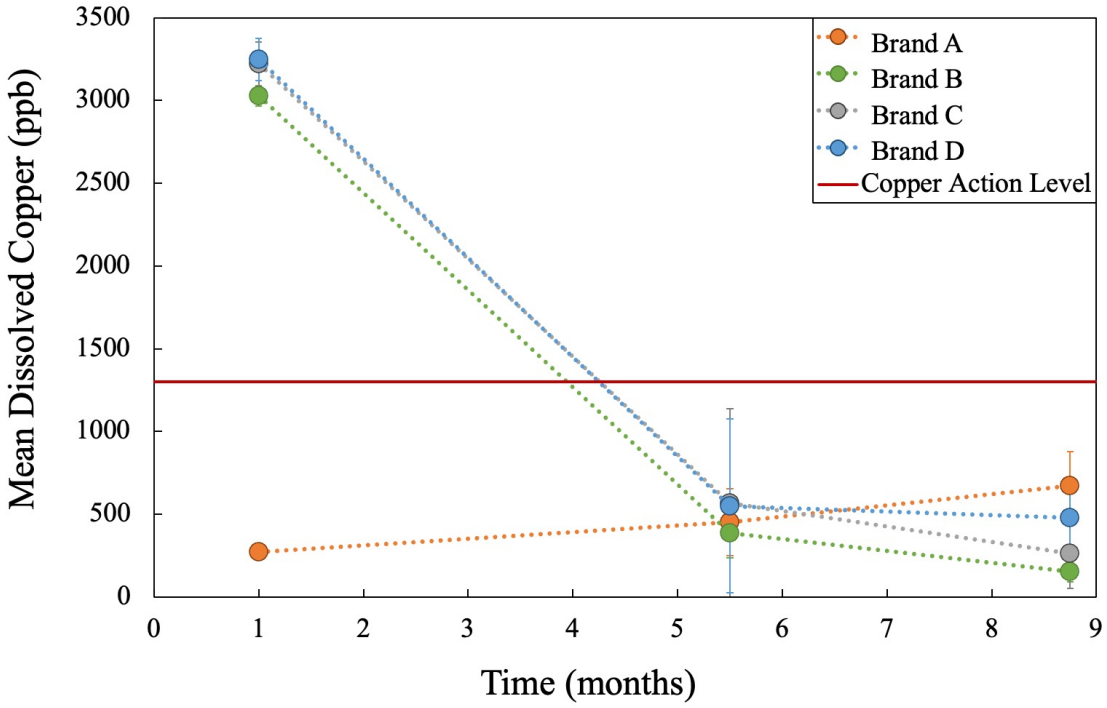


Figure 4.2: **Effects of tube manufacturer on cuprosolvency.**
Mean cuprosolvency observed over 8.75 months using pipe cuprosolvency tests: n=5 pipes per manufacturer, 4 manufacturers, at pH 8.25, 200 mg/L sulfate, and alkalinity of 500 mg/L as $CaCO_3$. Error bars represent one standard deviation.

These results call into question the long-term effectiveness of pH CCT and the "minimum" pH needed for a given tube type over a period of about 9 months or more. It is unclear if cuprosolvency in Brand A tube would eventually increase above the AL or if passivating

scales would form before copper levels exceeded unacceptable levels. These results could affect determination of "minimum" pH criteria, particularly at month 8.75, but could also give utilities a false sense of security if Brand A tube was used in cuprosolvency tests.

Variable effects of tube aging

Tube aging had significant, sometimes unexpected, effects that could complicate broad application of the "minimum" pH criteria for cuprosolvency control. Namely, several cases where copper did not decrease below the AL, even in 8.75 months, or even increased over time.

Cuprosolvency is generally expected to decrease with tube aging, eventually falling below the AL as passivating scales form, as occurred for Brands B, C, and D (Figure 4.2, pH 8.25 and 500 mg/L as $CaCO_3$). Significant ($p < 0.05$) decreases in cuprosolvency were observed in 60% of cases (month 1 to 8.75), however, copper remained above the AL for one quarter of these cases (Figure 4.3A). These results confirm that there are cases where protective scales may take more than 8.75 months to form, allowing high cuprosolvency to remain for a prolonged period even though copper might someday decrease to acceptable levels.

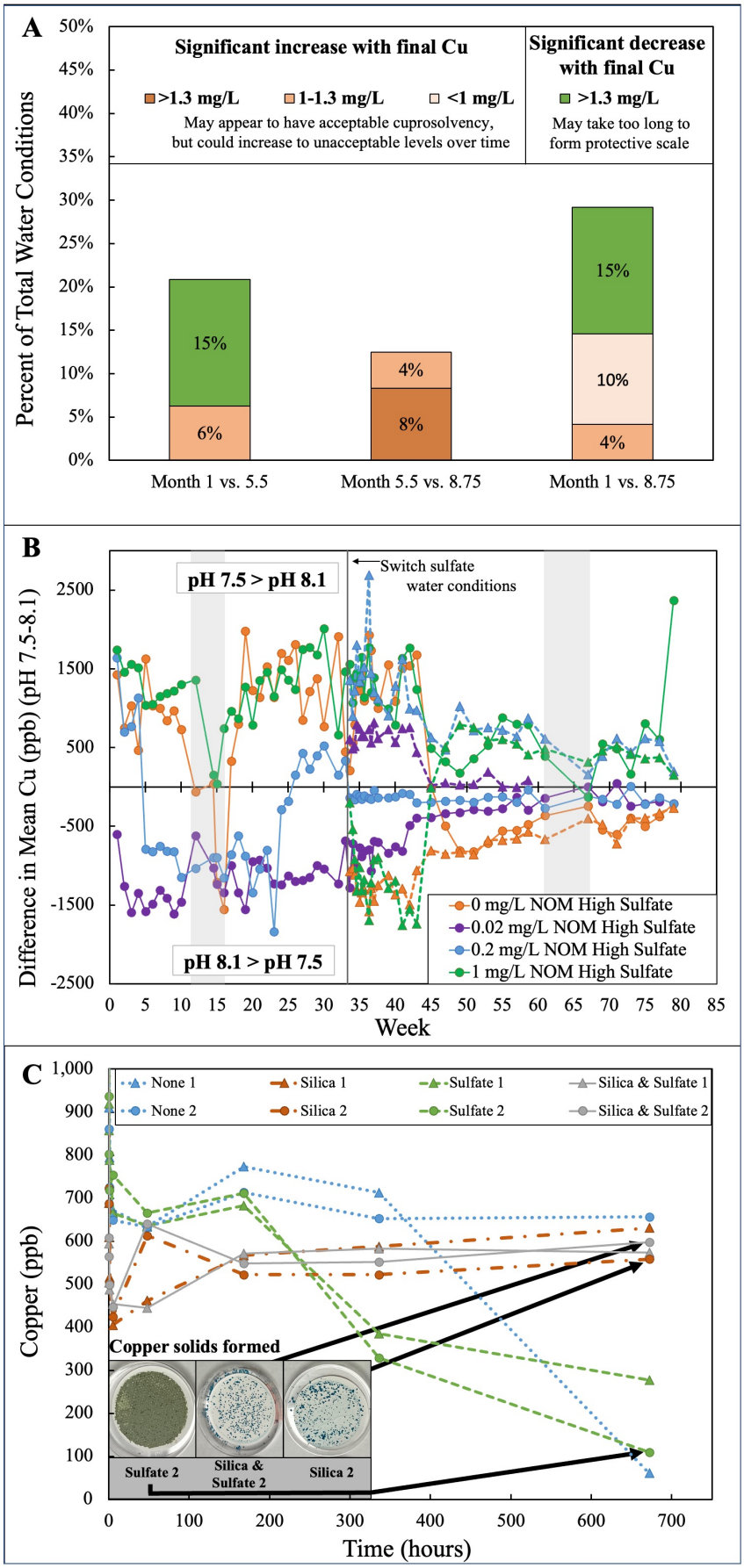

A
Significant increase with final Cu
>1.3 mg/L
1-1.3 mg/L
<1 mg/L
May appear to have acceptable cuprosolvency, but could increase to unacceptable levels over time
Significant decrease with final Cu
>1.3 mg/L
May take too long to form protective scale
Percent of Total Water Conditions
50%
45%
40%
35%
30%
25%
20%
15%
10%
5%
0%
15%
6%
4%
8%
15%
10%
4%
Month 1 vs. 5.5
Month 5.5 vs. 8.75
Month 1 vs. 8.75
B
pH 7.5 > pH 8.1
Switch sulfate water conditions
Difference in Mean Cu (ppb) (pH 7.5-8.1)
2500
1500
500
-500
-1500
-2500
pH 8.1 > pH 7.5
0 mg/L NOM High Sulfate
0.02 mg/L NOM High Sulfate
0.2 mg/L NOM High Sulfate
1 mg/L NOM High Sulfate
0 5 10 15 20 25 30 35 40 45 50 55 60 65 70 75 80 85
Week
C
None 1
Silica 1
Sulfate 1
Silica & Sulfate 1
None 2
Silica 2
Sulfate 2
Silica & Sulfate 2
Copper (ppb)
1,000
900
800
700
600
500
400
300
200
100
0
Copper solids formed
Sulfate 2
Silica & Sulfate 2
Silica 2
0 100 200 300 400 500 600 700
Time (hours)

Figure 4.3: **Challenges in broadly applying NDWAC criteria.**
A. Significant changes in copper over time. Significant differences ($p < 0.05$) in pipe cuprosolvency tests: n=5 pipes per water condition per manufacturer, 4 manufacturers, 3 pH values for each of 4 alkalinities.
B. Higher soluble copper observed at higher pH for some waters. Pipe cuprosolvency tests (n=5 pipes per condition) utilized waters with 250 mg/L as $CaCO_3$ alkalinity, 200 mg/L sulfate and various NOM concentrations (0-2 mg/L Fulvic acid) at pH 7.5 and pH 8.8. After week 33, the three pipes with highest soluble copper for each condition were switched to no sulfate.
C. Other water quality parameters may affect cuprosolvency: silica and sulfate. Fresh copper solids cuprosolvency tests using 0 or 0.5 mM sulfate and 0 or 8.5 mg/L silica on cuprosolvency of copper solids (0.5 mM cupric nitrate) in the presence of 1mM $NaNO_3$ and 250 mg/L as $CaCO_3$ alkalinity at pH 8.8. Inset shows variability in resulting solids.

Contradicting expectations, increases in cuprosolvency were also observed in a few cases (Figure 4.3A), calling into question the basic premise of the "minimum" pH criteria that copper would remain constant or decrease over time. Significant ($p < 0.05$) increases in copper concentration were observed in 14% of cases (month 1 to 8.75) with 4% having final copper concentrations above 1,000 ppb. Significant increases in copper were also observed for 12% of cases comparing month 5.5 to 8.7, where all had final copper above 1,000 ppb and 8% had copper above the AL. These exceptional cases demonstrate the potential for increased cuprosolvency over time. This was observed in the case of Brand A (Figure 4.2), where it is unclear if cuprosolvency will remain at acceptable levels or increase above the AL over time.

This variability in aging could affect development and application of "minimum" pH criteria. Regression criteria using the highest values from relatively new pipe are expected to yield conservative "worst-case" cuprosolvency estimates. Because copper concentrations did not always decrease or stabilize over time, it is unclear how longer aging times would affect regression criteria and therefore, if criteria represent the true worst-case. For example, variability was observed in regression criteria developed after 4, 22, and 35 weeks of

aging (Figure 4.1B), with criteria after 22 weeks resulting in markedly lower "minimum" pH criteria at high alkalinity than after other aging times. These results demonstrate several challenges in developing criteria and applying utility tests using copper tube cuprosolvency tests. Namely, consideration of whether the chosen duration for criteria development and/or utility testing is representative of long-term levels and if a stable scale will occur within a time frame utilities/residents deem acceptable.

Unexpected effects of pH on cuprosolvency

In contrast to the expected inverse relationship between soluble copper and pH, several conditions resulted in higher sustained cuprosolvency at higher pH. Anomalous behavior was observed in initial copper tube trials, where mean copper at pH 7.5 exceeded that at pH 7.25 (100 mg/L as $CaCO_3$ alkalinity) and concentrations at pH 7.75 exceeded those at pH 7.5 (200 mg/L as $CaCO_3$ alkalinity) for weeks 22-35. Similar anomalous behavior was observed in low NOM conditions (0.02-0.2 mg/L), where higher pH (8.1) yielded higher cuprosolvency than pH 7.5 (see negative differences in mean copper between pH 7.5 and pH 8.1 in Figure 3B). Results confirmed that this anomalous behavior was not caused by release of particulate copper, because particulates represented less than 15% of total copper when NOM was less than 1 mg/L at 17 and 22 weeks of pipe aging.

These results directly demonstrate several cases where increasing the pH could actually make cuprosolvency worse, at least within the time frame tested. Prior research also exhibited higher cuprosolvency at higher pH (pH 8.0 vs. 7.5) in early stages of cupric ion precipitation tests, however long term cuprosolvency followed the expected trends.14 Even though it is possible that a low solubility scale could eventually form in our waters, results indicate that the "minimum" pH criteria cannot work for all waters in less than 79 weeks.

Effects of other water quality parameters

In addition to the various factors described above, additional water quality parameters such as NOM, sulfate, and silica concentrations can affect cuprosolvency. These factors may affect the utility of the "minimum" pH criteria, yet they were not considered in the simplified NDWAC approach. We discuss predicted and observed effects of each parameter in the sections that follow.

NOM. Cuprosolvency testing in lab and field data previously revealed complex cuprosolvency responses to NOM, including a threshold type behavior with increasing NOM, causing a dramatic increase in cuprosolvency above 0.1- 0.25 mg/L as TOC or DOC.[44, 111, 179] Our study experimentally tested a much wider range of NOM under controlled conditions.

In this more detailed study, the presence of extremely low NOM concentrations (0.02-0.2 mg/L as fulvic acid, corresponding to 0.01 - 0.11 mg/L as C or up to 0.074 UV254 absorbance) appeared to reduce cuprosolvency, whereas higher NOM (0.5-2 mg/L as fulvic acid, corresponding to 0.27 - 1.16 mg/L as C or 0.15 - 0.52 UV254 absorbance) yielded higher cuprosolvency as expected (Figure C.3). Generally, cuprosolvency in low NOM conditions dropped and stayed below the AL earlier (pH 7.5, 0.02-0.2 mg/L NOM by 6 weeks; pH 8.1 0.02 mg/L NOM after 35 weeks, 0.2 mg/L NOM after 24 weeks) than higher NOM conditions (0.5-2 mg/L NOM), which had sustained cuprosolvency above the AL from 1,720 to 3,420 ppb after 33 to 78 weeks (1 mg/L NOM) of tube aging. But the surprising result was that the condition with no NOM, which may not occur in natural waters, also had higher long-term cuprosolvency above the AL, refuting a simple NOM threshold effect. Copper values in waters with no NOM did eventually drop and stay consistently below the AL after 45 (pH 7.5) to 71 (pH 8.1) weeks, indicating that long-term data may be in agreement with the literature. The effects of NOM were less pronounced at higher pH, possibly due to lower

initial cuprosolvency, with the maximum week 1 copper value at pH 8.1 about half that at pH 7.5.

Results also indicate complications with determination of an NOM concentration that constitutes a "worst" case aggressive water. A relatively high 0.2 mg/L NOM as fulvic acid concentration, expected to be above the reported NOM threshold level, was chosen to develop conservative "minimum" pH criteria. However, because somewhat higher cuprosolvency was observed at NOM concentrations above 0.2 mg/L, the "minimum" pH criteria may not be conservative enough for waters with higher NOM. Further, results indicate that the "minimum" pH criteria may not be applicably for extremely low NOM, which also resulted in relatively high cuprosolvency for about a year of tube aging.

Silica and Sulfate. Prior research indicated that silica and sulfate could also affect cuprosolvency and any framework neglecting their impact might be incomplete. The effects of sulfate and silica were tested using a faster and simpler fresh solids cuprosolvency method similar to that in Kriss et al.[181] Sulfate exhibited mixed effects on cuprosolvency. In copper tube testing, there was no apparent effect of reducing sulfate in 33-week aged pipes (200 mg/L to 0 mg/L). However, in fresh solids tests the presence of sulfate resulted in lower initial cuprosolvency (up to 336 hours) than in conditions with silica or no sulfate (Figure 4.3C). A similar initial benefit, but potential long-term detriment in cuprosolvency was observed with added silica, regardless of if sulfate was present. Conditions with silica resulted in higher final cuprosolvency than conditions with only sulfate (559 to 631 ppb vs. 109 to 277 ppb after 672 hours) and poorly crystalline solids with a discrete particle appearance, instead of crystalline solids with a more uniform appearance (Figure 4.3C, Section S1). The initial benefit, but potential long-term determent is similar to what was observed with orthophosphate addition and could be indicative of faster formation, in the presence of sulfate and/or silica, of a solid with intermediate solubility between the initial cupric hydroxide

and the malachite which may eventually form.[106, 113, 181, 264, 304] These results also indicate that the presence of silica may result in higher long term cuprosolvency, potentially via crystal poisoning or effects on the structure of scale formed, and that these effects may overtake the effects of sulfate. A deeper understanding of how these and other water quality parameters affect cuprosolvency and scale formation is needed to develop more widely applicable predictive models such as "minimum" pH criteria. Indeed, the literature predicts that sulfate could be an important parameter affecting cuprosolvency in aged copper tube.[347]

Proposed utility framework for cuprosolvency mitigation

Even though LCR revisions do not require utilities to address issues with persistent elevated cuprosolvency in new plumbing, guidance for doing so is still needed for proactive utilities, residents, and building managers attempting to deal with these concerns voluntarily.[27, 30] Our framework (Figure 4.4) updates guidance proposed in previous work to account for the many factors that may complicate broad application of "minimum" pH criteria without vetting it in each particular water.[15, 181] In this framework, a utility can choose to address suspected cuprosolvency systemwide via orthophosphate addition or adjusting water pH according to the "minimum" dose guidance. If a utility adjusts these parameters or wishes to demonstrate that their water is not aggressive to new copper, they can undertake bench scale or in-home cuprosolvency testing to verify effective cuprosolvency control. Finally, if utilities are unwilling or unable to address cuprosolvency issues via systemwide CCT and cuprosolvency testing, they can consider providing guidance to aid residents in implementing appropriate residential level interventions such as long-term avoidance (pipe replacement, NSF 53 certified water filters) or residence-level interventions such as whole-house pH adjustment.

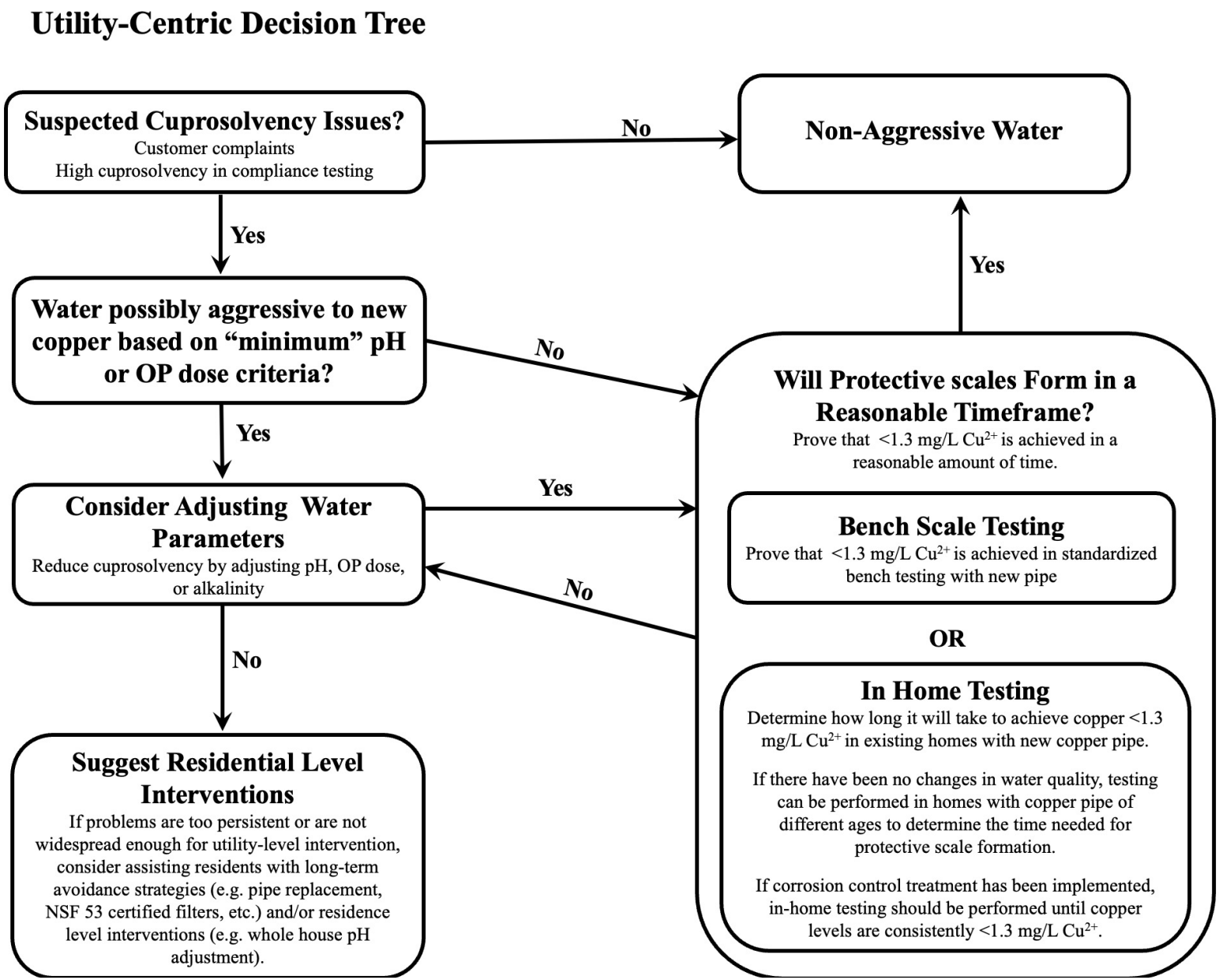


Figure 4.4: **Utility centric decision-making framework to address cuprosolvency issues.**

4.4 Implications

This work sought to test guidance suggested by the NDWAC Consensus Statement, that could be broadly applied to address issues with high cuprosolvency waters.[15] The underlying assumption of that approach is that, no matter how corrosive a water is, the pH can be increased high enough to cause cuprosolvency below 1.3 mg/L in a reasonable length of time. The literature predicts cuprosolvency reductions with increasing pH.[44, 98, 114, 155, 304] Previous work indicated that a similar approach could work for orthophosphate-based corrosion control, which was particularly advantageous because the system reached a pseudo steady state quickly, allowing a response to be observed within weeks.[181] However, this work demonstrates that it may take years or decades to observe cuprosolvency reductions with the "minimum" pH corrosion control approach. It is too simplistic and cannot be broadly applied across all water chemistries. In some of the "worst-case" waters tested herein, a relatively high pH of 8.1 was not successful in yielding acceptable cuprosolvency even after 1.5 years of aging.

Our work and the literature indicate that the formation and aging of protective scales is complicated and generally poorly understood. Observed cuprosolvency depends on a combination of pipe aging, which could occur in time frames ranging from seconds to decades, and the complicated interactions of a variety of water quality parameters.[44, 206] Many of these parameters act via competing mechanisms in themselves, such as increasing carbonate which exhibits complexation and pH stabilization effects, while also contributing to the formation of low solubility malachite solids.[98, 114, 302, 304, 347] It becomes increasingly complicated to predict net effects of combinations of water quality parameters which may have competing effects on cuprosolvency. For example, increasing pH can decrease cuprosolvency via formation of low solubility cupric compounds, while increasing NOM, sulfate, or silica can

increase cuprosolvency through complexation (NOM) and interfering with formation of passivating scales.[44, 98, 106, 111, 113, 114, 155, 179, 264, 304, 347] Further, variability in the effects of various water quality parameters have been observed in newer vs. aged pipes.[346] These modeling challenges are exacerbated by limited utility tracking and thermodynamic information for some of these components.[304] Overall, a better understanding of the interdependencies of various water quality parameters on cuprosolvency is needed in order to develop more broadly applicable cuprosolvency models.

Guidance is still needed to address cuprosolvency issues in drinking water, especially since LCR sampling is not representative of new copper plumbing most likely to have high cuprosolvency.[44] Although the minimum" orthophosphate guidance developed in previous work may be effective, the "minimum" pH criteria may not be able to ensure that waters are "non-aggressive" to new copper.[181] The most foolproof method to determine if waters yield acceptable cuprosolvency would be sampling in newer homes throughout the distribution system. If there have been no major changes in source water quality over time, sampling homes with copper tube of varying ages could be used to determine if a protective scale formed in a reasonable time frame, and consumers could be informed of typical "wait" times for aging in the system. However, if water quality is not constant across the system or if water quality has changed over time, utilities may need to perform longitudinal monitoring in a set of homes to determine how long it would take to form passivating scales. Further, utilities need to continue monitoring and adapting treatment as water chemistry changes continue to occur, such as those resulting from climate change, changes in air quality and water treatments, and variations in road salt use or water source.[40, 41, 261]

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Chapter 5

Guidance for Residents Addressing Copper Problems in Drinking Water: Opportunities and Challenges

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Abstract

Residents and their pets may experience aesthetic and health concerns resulting from elevated copper in their drinking water. The United States Environmental Protection Agency Lead and Copper Rule focuses on addressing systemwide corrosion issues, but gaps in the rule leave some municipal water consumers and residents with private well water vulnerable to high cuprosolvency. We developed guidance to aid residents in understanding, detecting, and addressing cuprosolvency issues in their drinking water. Three types of at-home test kits for copper and one for pH were determined to be accurate enough (R^2 >0.9 (lab) and >0.7 (field)) to detect concerns related to high cuprosolvency and guide intervention options. Case study results indicate that, although water treatments such as increasing pH on-site may be effective, long-term treatment (>36 weeks or permanently) may be needed to maintain reductions in cuprosolvency.

5.1 Introduction

Elevated copper in drinking water can be derived from corrosion of copper plumbing. High cuprosolvency, i.e. copper release to water, is expected in new plumbing and may persist for days, months, years, or decades until a low-solubility mineral coating, or "protective scale," forms inside the pipes. Whether this mineral coating will form and when, depends on water chemistry.[44, 106, 114, 179, 185, 304, 339] Residents with cuprosolvency issues may experience aesthetic concerns like blue water and fixture staining, as well as health effects such as acute gastrointestinal distress, liver and kidney issues, and more serious effects for certain sensitive populations (e.g. Wilson's disease) and some pets.[8, 115, 304, 329] Other manifestations of copper corrosion such as pitting, wall thinning, and erosion corrosion can be problematic, causing leaks and pipe failure, but are not the focus of this paper.[284] To address systemwide cuprosolvency issues, the United States Environmental Protection Agency (US EPA) Lead and Copper Rule (LCR) set an action level (AL) of 1.3 mg/L of copper in municipal systems. Copper concentrations for more than 90% of homes in LCR sampling must be below this AL, or utilities need to notify the public and implement corrosion control treatment (CCT).[8, 44, 49, 339] In some cases, even when utilities implement CCT, elevated cuprosolvency may persist indefinitely, as was observed for a utility adding polyphosphate corrosion inhibitors.[58]

The LCR may leave some residents unprotected from elevated copper in drinking water. Because the LCR uses a 90^{th} percentile AL, up to 10% of residents could be vulnerable to elevated copper above the action level. In addition, homes sampled under this rule are generally not representative of the worst cuprosolvency issues, because the rule prioritizes sampling of older homes at highest risk of lead, rather than the homes with new copper pipes which tend to have higher copper.[44, 339] Therefore, LCR sampling is likely to underestimate

potential cuprosolvency issues for some new homes, and the extent of this underestimation keeps increasing as homes in the LCR sampling pool age. Finally, because the LCR regulates water quality of homes in municipal systems, elevated copper exposure from drinking water via private wells and non-residential buildings are not addressed.[49, 99, 304, 339] These gaps leave all private well owners and many residents and non-residential building owners with municipal water, at risk of cuprosolvency issues in their drinking water.[44, 115, 270]

Consumer-centric tools and guidance are therefore needed to help address elevated cuprosolvency. Guidance was previously developed to help utilities address cuprosolvency based on key water quality parameters such as pH, alkalinity, and orthophosphate dose.[15, 180, 181] That guidance serves as a starting point for consumers, but needs to be modified to consider sampling methods and treatments appropriate for residential use. Some at-home tests for lead and chlorine in drinking water were relatively accurate and appropriate for citizen science use,[182, 285] suggesting that similar tests may be promising for measurement of copper and water quality parameters needed to guide consumer-centric strategies.

Ideally a hierarchy of treatment strategies could be proposed with a preference for effective, shorter-term, less costly strategies (Table 5.1). One-time and short-term site specific treatments that could facilitate permanent formation of a low-solubility (i.e. protective) scale would be ideal for residential applications, such as the granular activated carbon removal of organic matter suggested by Arnold et al. or anion exchange treatment.[44] However, researchers have noted difficulties applying these types of laboratory cuprosolvency results to the field, indicating a need for further study to determine if and when they would be successful in real systems.[106, 304] Therefore, more established longer-term or permanent water chemistry adjustments (i.e., dosing phosphate, raising pH) may be needed to reduce cuprosolvency. However, these treatments may present too much expense and effort (installation, maintenance, reagents) so residents may prefer to take other steps such as expensive

one-time replumbing with plastic pipe or avoiding copper exposure by flushing, use of NSF 53 certified filters, or drinking bottled water.

Table 5.1: **Hierarchy of potential residential interventions to address copper in drinking water**
*NSF/ANSI Certified point of use filters[237] and reverse osmosis treatments[238]
GAC refers to Granular Activated Carbon filtration
Approximate costs are as follows:
$ indicates less than $50 initial capital cost or maintenance per year
$$ indicates $300 - $1000 initial capital cost or maintenance per year
$$$ indicates more than $1000 initial capital cost or maintenance per year

Intervention	Confidence of Resolution		Initial Capital Cost	Ongoing Maintenance	Additional Considerations
	Consumption And Cooking	Household Water			
Replumb with Plastic or Stainless Steel	Certain		$$$	0	High cost and construction in home
Bottled Water	Certain	No effect	$	$$	No effect on fixture staining or green hair
Point of Use Tap or Pitcher Filter*	Likely	No effect	$	$	At present no tap or pitcher filters are NSF/ANSI 53 certified for copper removal
Point of Use Reverse Osmosis*	Certain	No effect	$$	$	NSF/ANSI 58 certified for copper removal
Whole House Orthophosphate Addition	High		$$	$	Active chemical dosing likely required.
Whole House pH adjustment	High Dependent on water chemistry		$	$ or $$	Active chemical dosing of caustic, or passive limestone/soda ash contactors
Whole House GAC, Reverse Osmosis, Ion Exchange	Unproven/Unknown Dependent on water chemistry		$$	$$	More research is needed

This study aims to develop residential guidance to detect and address elevated cuprosolvency in drinking water. A preliminary decision framework was adapted from prior research to adjust testing and intervention strategies to be appropriate for use in individual residences. This framework could be used either by individual residents or adapted by water authorities (i.e. utilities, health departments, cooperative extension programs) to provide location-specific guidance accounting for concerns/conditions in specific water systems or

water sources. The laboratory and field accuracy of inexpensive at-home test kits were evaluated for providing data to navigate the framework. We also demonstrated the efficacy of potential interventions in both lab and the field studies and use of a simple diagnostic cuprosolvency test to rule out grounding or pipe defects. Finally, we illustrate use of the evolving framework via case studies with two residents dealing with cuprosolvency issues.

5.2 Materials and Methods

5.2.1 At-home Test Kit Testing

At home test kits for copper and pH were tested according to manufacturer instructions to determine their accuracy in the laboratory (1 participant, blind sampling, n=5 tests per condition) across a range of pH and copper concentrations. Test kits included color tile strips and liquid-based color change kits for both pH and copper and a low-cost field colorimeter for copper. Laboratory test waters were comprised of deionized water adjusted to target pH values (6.0 to 8.8) or with cupric nitrate (0 to 5 mg/L copper) and performed within one hour of copper addition. Field accuracy of these test kits was determined via residential sampling performed by Resident B.

5.2.2 Residential Water Sampling and Testing

Resident A collected water samples (250 mL) at two locations (kitchen tap and hose bib) in their home, which was replumbed with copper pipes in 2017. For comparison a tap in a next-door building with copper pipes >40 years old was also sampled. One sample was collected after flushing approximately 5 gallons of water (5 minutes at kitchen tap), another

was collected immediately upon opening the tap after a prescribed stagnation time (4 hours during the day, 10 hours at night, and up to 24 hours in subsequent tests), and a third after 20 seconds of flushing post-stagnation. However, because the next-door building has daytime occupants, daytime stagnation goals could not be ensured. Resident A determined electric current on pipes according to a method suggested by a technical expert at a major multimeter manufacturer, whereby the current was measured between the hose bib or the water meter and an electrical connection to a grounded pipe segment in the yard away from the plumbing. An onsite cuprosolvency test was performed using 8.5-inch copper pipe segments (n=5) filled with water from the residence and closed with silicone stoppers. The pipe segments were filled with water collected from the residence just after the 5-gallon flushing step and water was collected for copper analysis after the 10-hour overnight stagnation period. This allowed for direct comparison of results from the home pipes potentially affected by current, and the test pipes not connected to the home plumbing and therefore having no current.

In-home sampling was also performed by Resident B in a nearby residence connected to the same municipal water system. Their home was renovated from 2014-2017, including the installation of new copper pipes. Resident B collected three samples (250 mL) weekly each from a tap in their home and in a guest house on their property from March of 2021 to May of 2022. Samples were collected after a 5 second flush to obtain water from copper plumbing and included samples collected immediately upon opening the tap, after 5 minutes of flushing, and after water was stagnant for 24 hours. Samples were analyzed in the field using citizen science testing with at-home test kits and with a calibrated field pH probe. Laboratory analysis was used to confirm copper concentrations. A whole house treatment system using soda-ash pH adjustment was installed by Resident B and periodically utilized to treat the water.

5.2.3 Water Treatment Testing

Laboratory cuprosolvency tests investigated the effects of utility-provide source waters and several water treatments. These tests followed a similar protocol to that of Kriss et al. (2023), however experiments were performed at ambient temperature.[180, 181] In brief, experiments used 8.5-inch copper pipe segments stoppered at one end with silicone stoppers (n=5 per condition), with water changes three times per week via a dump and fill protocol. Weekly composite samples and monthly individual pipe samples were collected and analyzed for copper. The local utility indicated that the resident likely received water from multiple sources, so one surface and two well source waters provided by the utility were tested. Additional water conditions tested include reverse osmosis treated water, and water sent by the resident that was treated using pH adjustment with soda ash (initial pH 7.5, pH 8.8 and pH 9.5), granular activated carbon filtration, and filtration using an anion exchange resin.

5.2.4 Analytical Methods

Standard methods were used to analyze base water quality parameters. Samples were analyzed for copper using atomic absorption spectroscopy (AAS, Perkin-Elmer 5100 PC AAS) via method 3111B or inductively coupled plasma-mass spectrometry (ICP-MS; Thermo Scientific iCAP RQ ICP-MS) via method 3125 B.[72] ICP-MS analysis was also used to determine concentrations of other metals, phosphorous, silica, and sulfate. Analyses using AAS and ICP-MS were performed after acidification of samples with 2% nitric acid for at least 16 hours.

5.3 Results and Discussion:

5.3.1 Residential Guidance Framework

Gaps in the LCR leave many residents vulnerable to persistent elevated copper in their drinking water, demonstrating the need for guidance to help them address cuprosolvency issues. Ideally the water coming into residents' homes would either be non-corrosive to new copper pipes, or utilities would treat it to render it non-corrosive across the entire distribution system. However, in many cases utilities are not required to implement corrosion control treatment under the LCR, or residents/building owners are solely responsible for their water quality (e.g. private wells).[8, 44, 49, 115, 339]

A guidance framework was developed to mitigate risks and help residents detect and address cuprosolvency issues (Figure 5.1). In this approach, residents would initially confirm that they have elevated copper in their drinking water. If their copper pipes are relatively new (<6 months old), they are advised to wait until pipes are 6 months old, allowing time for a protective scale to form. If high cuprosolvency persists for more than 6 months, residents may wish to perform simple at-home cuprosolvency tests to rule out factors such as installation and pipe manufacturing issues and improper grounding, which are often blamed for high cuprosolvency, even though they are rarely the cause of such problems in our experience.[184, 224] Residents can also consider adjusting the water quality through whole-house treatments to facilitate the formation of a protective scale. This includes using relatively simple off-the-shelf treatment systems with common corrosion control methods such as adjusting the pH of the water (i.e. via limestone contactor or adding soda ash) or adding orthophosphate corrosion inhibitors, with treatment targets guided by criteria from previous work.[15, 180, 181] Additional treatments such as filtration with granular active

carbon, might reduce cuprosolvency in some cases, but would require further research to determine their broader applicability.[44] Residents can always take steps to avoid ingesting water with elevated copper (flushing, NSF 53 certified filter, bottled water) while waiting for a protective scale to form, if they prefer not to use other interventions, or if other approaches are not successful. As a last resort replacing the copper pipes with stainless steel or plastic pipes can be considered. This approach may be helpful for individual residents, building managers, and proactive utilities in cases where cuprosolvency concerns can be addressed on a small-scale basis and are not widespread enough to warrant utility-wide corrosion control.

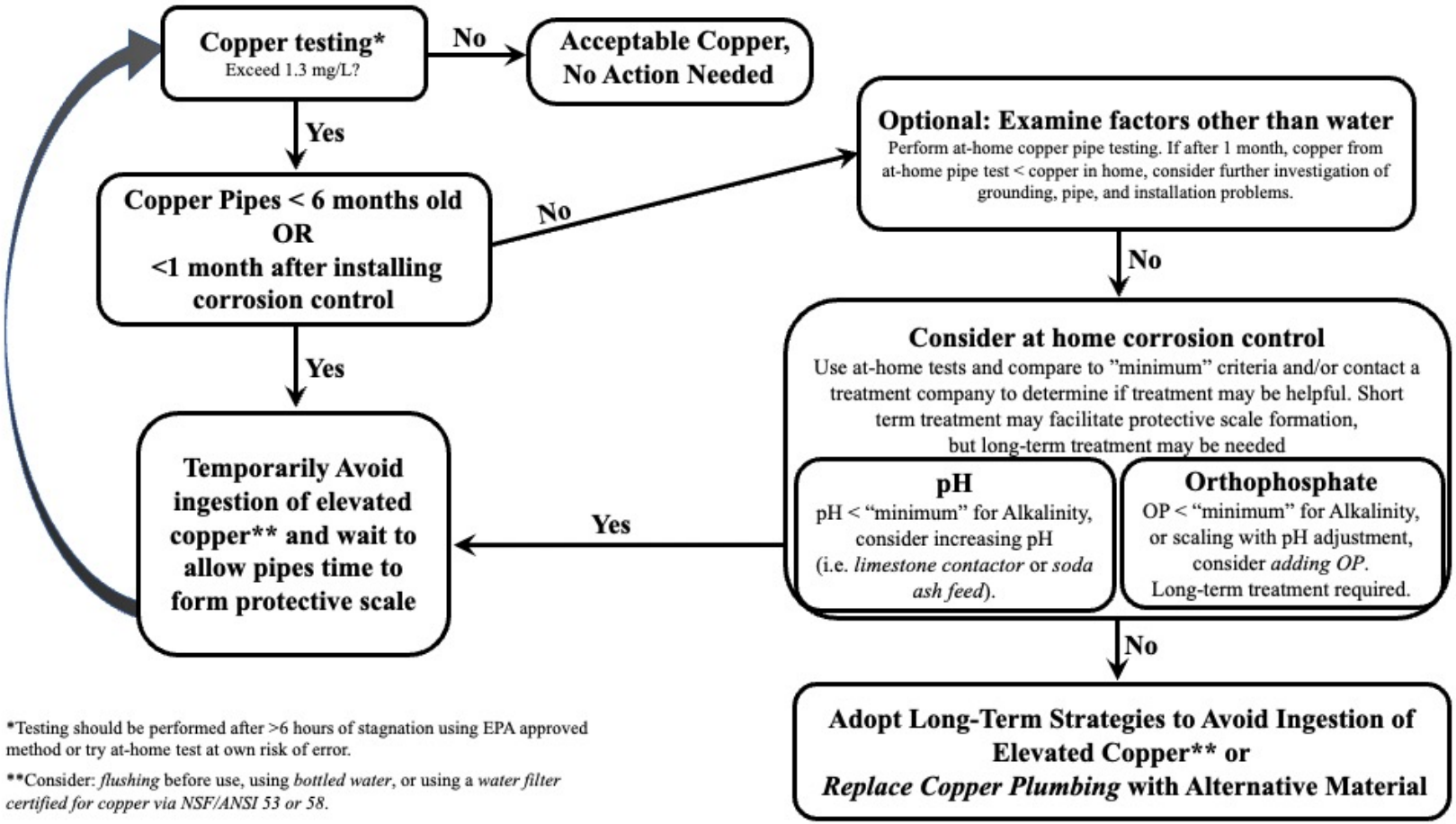


Figure 5.1: **Resident centric decision-making framework to address cuprosolvency issues.** "Minimum" criteria for potential corrosion control were determined in previous studies.[180, 181]

5.3.2 Accuracy of at-home copper and pH test kits

While consumers could always pay for sampling by a certified lab, inexpensive consumer-centric tools would be useful to help residents determine if they have elevated copper and to determine water quality parameters that could affect treatment recommendations. Many at-home tests for parameters like copper and pH are available for drinking water, pool, and aquarium uses. Three types of at-home tests for copper and two types of at-home tests for pH were evaluated in the laboratory and the field to determine their accuracy and general usability (Figure 5.2). Test kits included color tile strips and liquid tests for both parameters and a field colorimeter for copper detection. All test kits utilize color changes to indicate copper concentrations or pH values, but the field colorimeter gives a more precise digital readout.

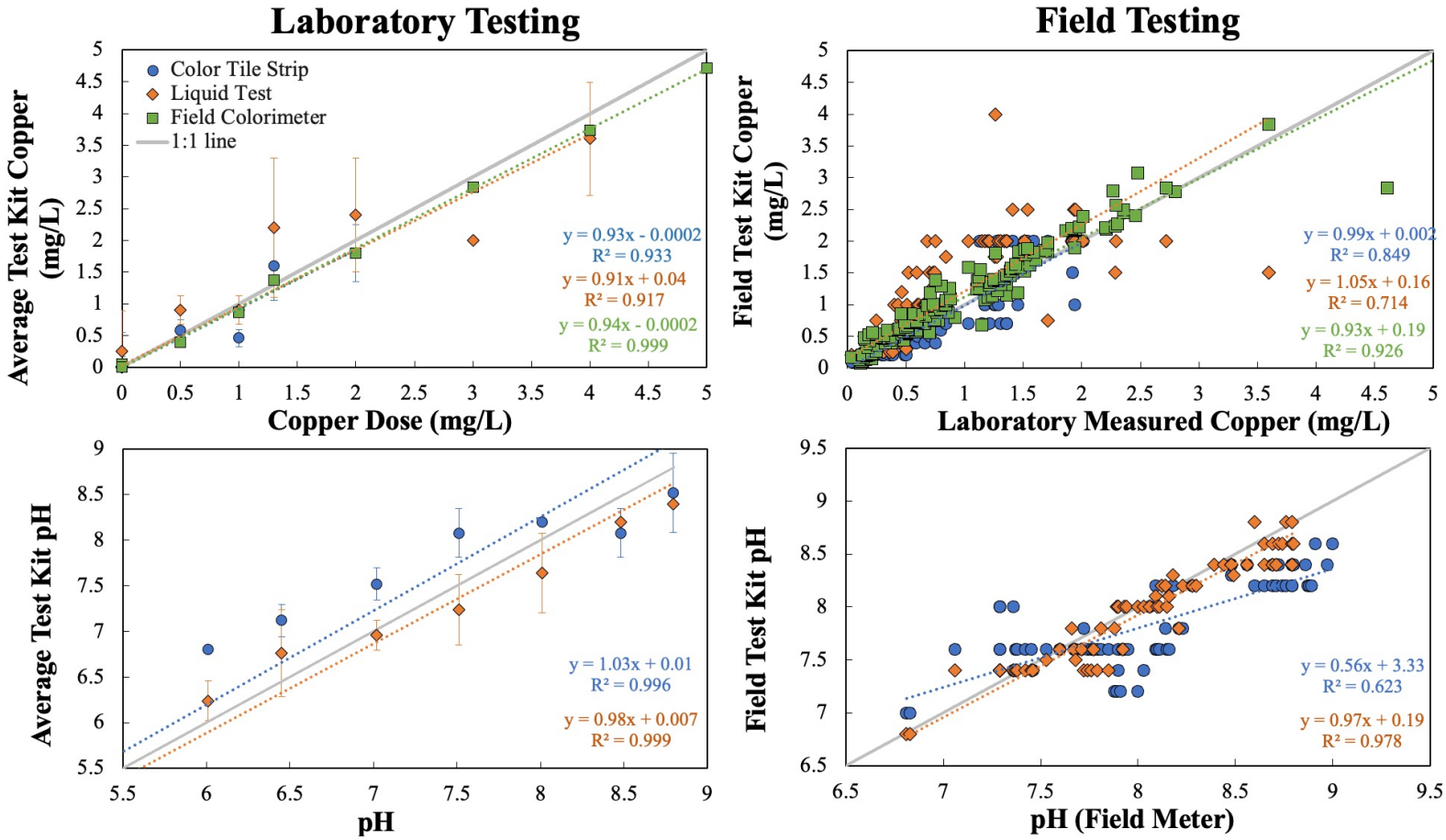


Figure 5.2: **Laboratory and field accuracy of at-home pH and copper tests.**
Laboratory and field accuracy of pH and copper at-home test kits (color tile, liquid, field colorimeter). Laboratory results were determined using a blind testing approach (n=5 replicates). Field results were measured by Resident B over the course of 1.5 years of in-home sampling. Actual values were determined via ICP-MS/MS laboratory analysis (copper) and a calibrated pH meter.

Results indicate that the field colorimeter is the most accurate option for relatively low-cost detection of copper in drinking water. The field colorimeter exhibited good accuracy in both the lab and the field ($R^2 > 0.93$), with relatively small variability (laboratory relative standard deviations 1-15%) and no false negative results (i.e. cases where the kit falsely indicated copper below the AL), making it the best option for copper detection. The liquid test was the least accurate (lab R^2 0.92, field R^2 0.71), and exhibited more variability (laboratory relative standard deviations 0-50%), but still low false negative rates of only 5%. Finally, even though the color tile strip was generally accurate in both the lab and the field (lab R^2 0.93, field R^2 0.85), it exhibited substantial variability (laboratory relative standard deviations 25-35%) and the highest false negative rate of 30%, making it the least reliable choice for detecting copper. These results demonstrate the inexpensive ($64, plus $0.50/test) field colorimeter is accurate enough to serve as a relatively low-cost initial screening tool to detect copper above the EPA AL and to potentially monitor progress as treatments are implemented.

Laboratory results indicate that both liquid and color tile strip pH tests are relatively accurate ($R^2 > 0.99$ and relative standard deviations $< 7\%$). However, the liquid test (R^2 0.978) was more accurate in the field than the color tile strip (R^2 0.623). Therefore, the liquid test is recommended for residential use and could be used in conjunction with the guidance framework to indicate if pH CCT could be a viable treatment strategy. Residents should still consider if more sophisticated tests are needed prior to investing in whole-house treatment strategies.

5.3.3 Citizen Science Case Studies

This section relates the experiences of two residents navigating the guidance framework while trying to address cuprosolvency problems in their homes. Both residents receive their water from the same municipal water system which did not detect systemwide cuprosolvency problems since at least 2013, (90^{th} percentile copper below 0.31 mg/L).[13] Despite this, these residents experienced prolonged issues with elevated cuprosolvency after installing new copper pipe during home renovations. Other residential buildings on the same water system also reported elevated copper in 2017, indicating that more widespread issues may be present than detected via LCR sampling.[149]

Resident A Experience Investigating Non-Water Factors

Resident A completed installation of new copper pipes in 2017. Two years later, their dog became ill and nearly died with symptoms later determined to be consistent with heavy metal poisoning, and then made a complete recovery after switching from tap to bottled water.[305] By September of 2020 the resident was notified that they had elevated copper (1.53 mg/L) in their water, which was found through routine LCR sampling. Even higher copper concentrations (2.45 mg/L) were found in follow up sampling.[359] This represents a case where the LCR sampling detected cuprosolvency problems. However, because the utilities aggregate 90^{th} percentile copper concentration was less than the AL, no corrosion control treatment or utility action was required (i.e., up to 10% of homes sampled can exceed 1.3 mg/L without taking action).

Authorities initially blamed high current due to improper grounding for this resident's elevated cuprosolvency. The current was allegedly so high in November of 2020 that the resident was advised to leave the premises due to shock hazard.[305, 359] In the next few

weeks, the resident worked with local water, gas/electric, and telecom utilities to address the issue. Even after multiple utility interventions, the stray current was reduced but not fully removed from the resident's pipes and copper levels remained above the AL, near 3 mg/L.[305] In December of 2020, Resident A installed a dielectric union to electrochemically disconnect their pipes from the copper service line which reduced the remaining voltage on the pipes from 30 volts to none.[359] But elevated copper persisted in the resident's home.

Having exhausted other avenues, the resident connected with the authors in fall of 2021 with concerns about elevated copper, still convinced it was due to a very low direct current on their pipes. We helped them measure copper in the water in their home with relatively new pipe (<5 years old). Results indicated much higher copper in the home (0.5-1.1 mg/L) than in a next-door building with approximately 40-year-old pipes (<0.1 mg/L), suggesting a protective scale had not yet formed in the resident's home. Flushing the water for 5 minutes at the kitchen tap (5 gallons) was effective at reducing copper to 0.1-0.2 mg/L.

In order to address the resident's concerns about any remaining current on their pipes, we helped the resident test copper concentrations when their water was in contact with brand new pipe segments without any electrical connection, as suggested in the decision framework (Figure 5.1). This revealed higher copper concentrations in the brand-new pipe segments with no current (0.98-1.31 mg/L) versus the 40-year-old grounded pipes in the home (0.50-0.96 mg/L) (Table D.1), indicating that the water alone could be responsible for the high copper. The resident also performed in-home sampling which yielded no consistent relationship between current and copper concentrations.

In the summer of 2022, because copper levels stayed high (1-2.4 mg/L), the resident installed a sacrificial anode in an attempt to address any remaining current issues. Over the course of two months, the resident installed, connected, disconnected, removed, and moved the anode, with no discernable effect on the copper concentrations in their water. Although this resident

did initially have grounding issues their more recent results are consistent with our experience that grounding is rarely the cause of elevated copper in water. In the end, the resident was relying on a reverse osmosis filter and considering replacing all their copper pipes. This demonstrates the struggles and frustrations of having individual residents navigating cuprosolvency issues.

Resident B Experience Investigating Water Quality Factors and Potential Treatments

Resident B's experience, summarized in Figure 5.3, began with a home renovation and installation of new copper pipes from 2014-2017. In the summer of 2020, more than 3 years after the renovation, the resident suspected cuprosolvency problems after noticing blue deposits in their icemaker and a green tint to their hair. Similar hair color changes and blue deposits were observed in several instances in the community 3 years earlier.[149] The resident used an at-home test which indicated copper concentrations above the AL (1.7 mg/L) in their water and confirmed elevated copper up to 3.88 mg/L via private laboratory testing in September and October of 2020. After hearing about the experience of Resident A, Resident B suspected that their dog, who got sick and died soon after the renovation, may have also been affected by heavy metal poisoning from elevated copper in their water.

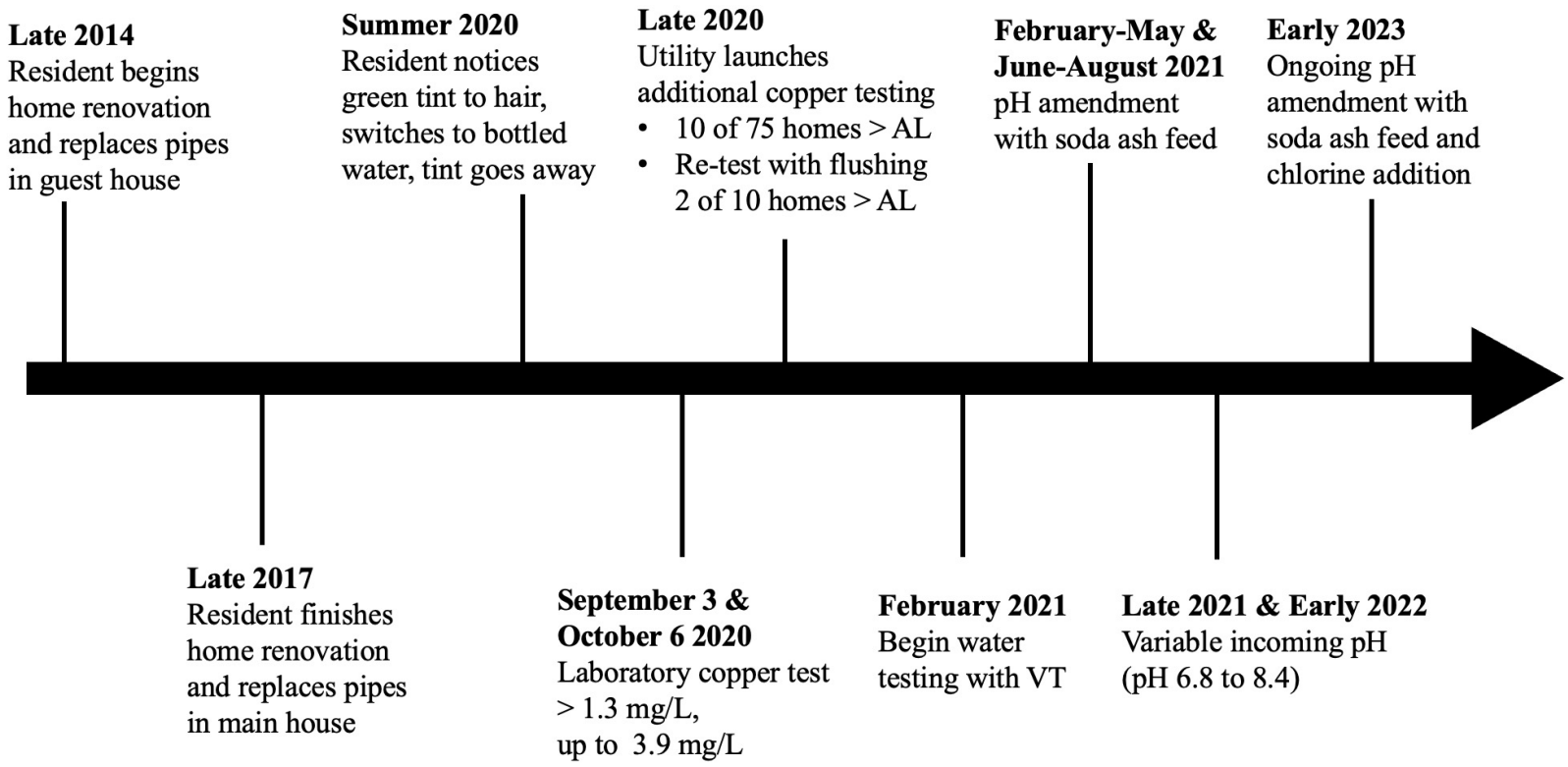


Figure 5.3: **Resident B Timeline.**
Timeline detailing pertinent events related to Resident B's installation of new copper pipes, determination of, and addressing cuprosolvency problems in their water.

Resident B first tried to address these issues in collaboration with their local water utility. In response to local reports of elevated cuprosolvency, the water utility performed additional non-LCR testing in late 2020, including testing water in Resident B's home.[305] They found copper above the AL in 10 of 75 homes tested, but only two homes still had copper above the action level after re-testing using a pre-flushing protocol. The utility asserted that any cuprosolvency problems were an isolated occurrence, and the residents were on their own to address any cuprosolvency issues. But the utility continued to cooperate with us by providing water and information when requested.

Resident B undertook a variety of steps to address the cuprosolvency issues. After initially discovering elevated copper, the resident switched to drinking bottled water, and installed a point of use reverse osmosis system, which alleviated their green hair issues. The resident then began investigating and implementing whole house treatment options. First, the resident bypassed the water softener for 10 days, hoping in vain to cause formation of protective scales. Copper concentrations remained high at 1.59 to 3.63 mg/L after treatment as measured by a private laboratory testing. The resident then worked with a local home water treatment company to install a whole house system to raise the water pH and monitored copper concentrations with an at-home colorimeter. A soda-ash feed was chosen instead of using a limestone contactor, which has easier maintenance, due to the relatively high dissolved inorganic carbon content in the water.[315]

The resident had a strong applied chemistry background during his career in industry. They contacted the Virginia Tech team in February of 2021 to gain a deeper understanding of these cuprosolvency issues. We helped the resident assess several factors and assisted with laboratory copper analysis. Tests indicated that sulfate reducing bacteria, which can increase cuprosolvency, were not present.[348] Further, tests indicated that short-term chlorination of the system via flushing or bypassing the water softener, which can sometimes promote

copper oxidation and aging, was not effective at reducing copper.[231] Testing also revealed that flushing the water for 5 minutes was not always sufficient to reduce copper below the action level, with concentrations up to 1.5 mg/L. Finally, we helped the resident select a target pH of 8.5 for pH adjustment with the soda ash feed. This pH was predicted to exceed the minimum criteria in the framework (Figure 5.1), and was therefore expected to yield a water that is non-aggressive to new copper.[15, 180]

Repeated in-home sampling revealed highly variable copper concentrations, which were frequently above the AL and up to 5.1 mg/L without any intervention (Figure 5.4). Continuously boosting the pH using the soda-ash feed was effective at quickly reducing cuprosolvency below the AL. However, when treatment was suspended copper concentrations increased back above the AL, indicating that a permanently protective scale did not form during either of the two 3-month periods of pH adjustment. During the first period of treatment, it was determined that the soda-ash had a polyphosphate additive, often used to prevent scaling and reported to complex copper, which could have affected potential protective scale formation during this period.[116, 206] The resident also noted initial difficulties maintaining the target pH, having to perform frequent adjustments to the system, which may have been caused by variations in the incoming water quality. Fluctuations in the incoming pH ranged from 6.8 to 8.4 when no treatment was being used (late 2021 to early 2022). The utility indicated that, due to the resident's location in the distribution system, they likely received a mixture of surface and well water, which could have contributed to these variations. Using Si and Cl measurements (via ICP-MS) as a tracer indicated that about 25% of the resident's water was from wells and 75% from surface water at the time tested. These results highlight some challenges residents may face when attempting to implement whole house treatment strategies.

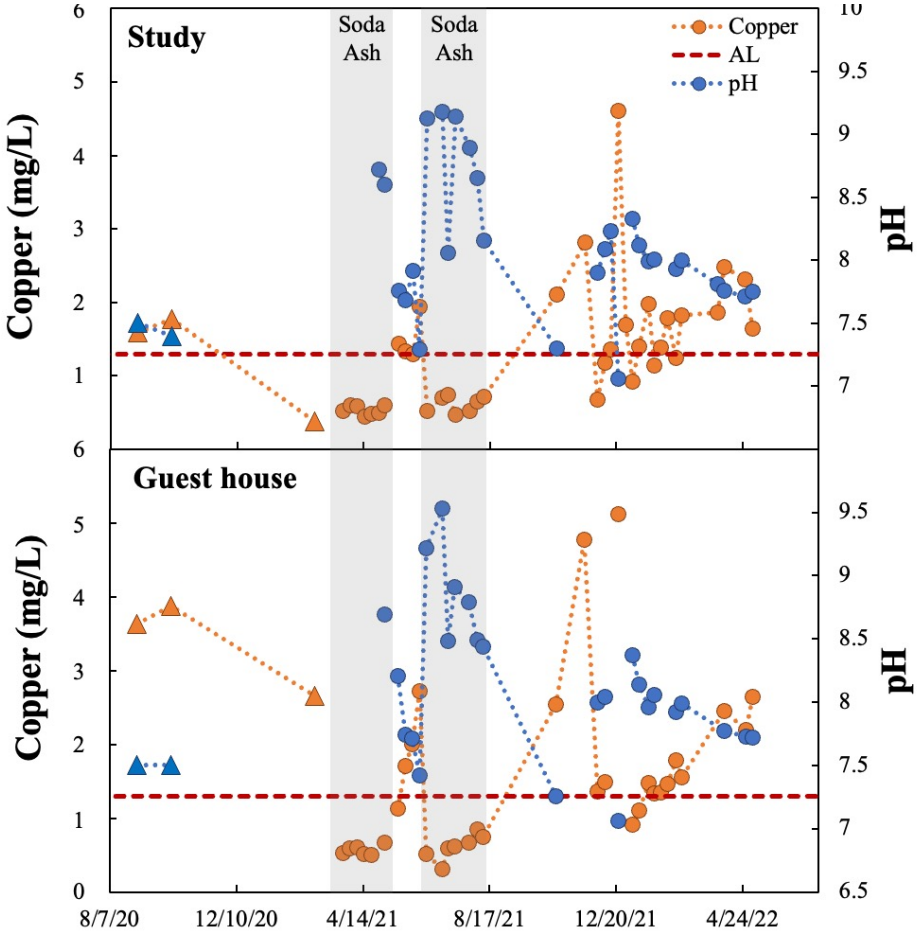


Figure 5.4: **In home testing/intervention.**
Copper and pH measurements from the home of Resident B at two locations. Historical data from testing in the home are represented by triangles. Circles represent first draw samples collected in this study after 24 hours of stagnation.

Laboratory pipe cuprosolvency studies were performed using Resident B's water as well as water from the local utility to determine potential causes and effective treatments for the observed high cuprosolvency (Figure 5.5). Results indicate that Resident B's water (pH 7.5) generally resulted in intermediate cuprosolvency between utility surface and well water sources, consistent with reports from the utility that Resident B receives a mixture of these waters. Cuprosolvency in these waters remained relatively high (>0.9 mg/L) within the 36

weeks of testing, with the resident's water (>1.6 mg/L) and the surface water source (all but one sample >2.3 mg/L) remaining almost always above the AL. Even though the utility was not exceeding the AL and was therefore not in violation of the LCR, these results confirmed that a low solubility protective scale did not form in new pipes exposed to their water for up to 36 weeks—even without any grounding or other dubious causes of cuprosolvency. This is consistent with the elevated cuprosolvency observed in both Resident A and B's homes even after 5 years of aging as well as elevated cuprosolvency observed in condo buildings in the community after 5-10 years of pipe aging.[149]

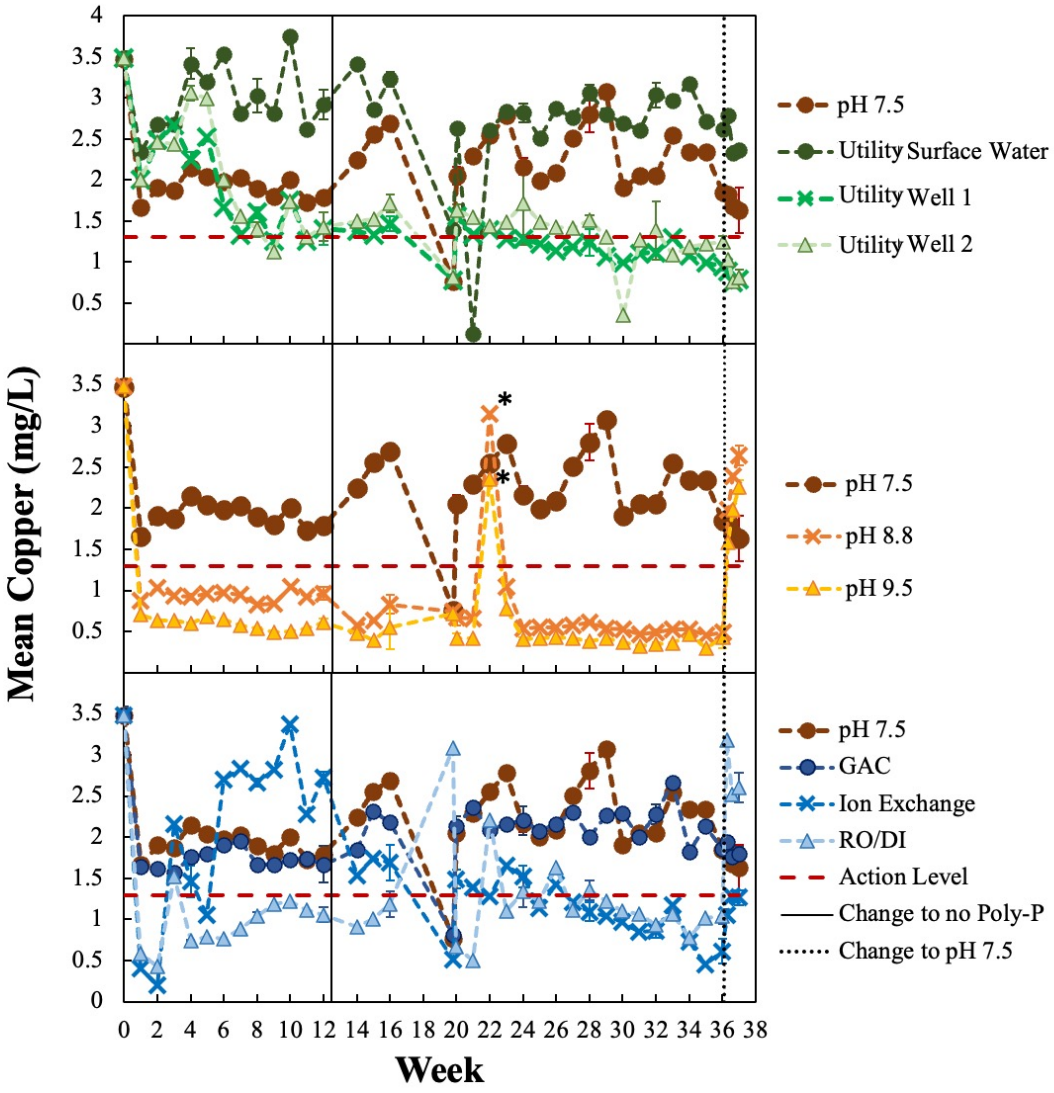


Figure 5.5: **Lab pipe tests with interventions.**
Laboratory cuprosolvency tests using copper pipe (n=5) and Resident B's water (pH 7.5), Resident B's treated water, or surface or well water from the local utility. Water treatments include filtration with granular activated carbon or anion exchange resin, reverse osmosis/deionized water, and pH adjustment using soda ash to pH 8.8 or 9.5. All waters had polyphosphate prior to week 13, except water from the local utility and RO/DI water. Polyphosphate was removed by the ion exchange treatment. All waters were changed to Resident B's control water (pH 7.5) after week 36. Error bars represent standard deviations for full sampling events.
* pH 8.8 and 9.5 waters were adjusted to pH 7.5 for week 22.

In order to address the cuprosolvency issues, potential treatments for the resident were evaluated at Virginia Tech using pipe cuprosolvency tests. The most effective treatment tested was the addition of soda-ash to increase the pH to pH 8.8 and 9.5, above the predicted "minimum" threshold value. Upon increasing the water pH, copper concentrations quickly fell below the AL to as low as 0.3 mg/L. However, drastic increases in cuprosolvency of 1.1-2.5 mg/L were observed when the pH was returned to the resident's original pH 7.5 water, even after 22 or 36 weeks of treatment, indicating that a permanent protective scale had not formed. Reverse Osmosis pre-treatment yielded cuprosolvency reductions from 0.3 to 1.9 mg/L in comparison to no treatment (pH 7.5 water), however copper concentrations remained relatively high near the action level for the majority of the test. Anion exchange filtration resulted in cuprosolvency consistently below the AL after 27 weeks of treatment, with concentrations as low as 0.5 mg/L. Similar to pH treatment, increases in cuprosolvency of 1.5-2.1 mg/L and 0.5-0.7 mg/L, respectively, were observed for both reverse osmosis and anion exchange treated waters when treatment was suspended. Granular activated carbon filtration to remove natural organic matter was the least effective treatment, exhibiting no apparent effect on cuprosolvency in Resident B's water.

5.4 Implications

Gaps in the LCR leave many residents at risk of elevated copper in their drinking water.[44, 49, 99, 115, 270, 304, 339] The case studies described above represent one exemplary case where the utility was in compliance with the LCR, but their water was aggressive to new copper plumbing, yielding elevated cuprosolvency for a prolonged period of time. Even though there are multiple reports of elevated cuprosolvency, including our community partners and other residences reporting elevated copper for 5-10 years following installation of new pipes,

the utility bore no responsibility for addressing these problems.[149]

It would be ideal for local authorities (e.g. utilities, health departments, cooperative extension programs) to take the lead on developing treatment and intervention guidance based on their specific water sources. In this case, the guidance herein could serve as a starting point, with the local authority adapting it to account for local considerations and acting as a centralized and reliable information source to help residents navigate cuprosolvency problems. This would leverage existing expertise and specialized knowledge of local and/or municipal water quality and appropriate treatments. However, in cases where local authorities abstain from assisting, residents are left largely on their own to address cuprosolvency issues. This puts a huge and undue burden on consumers, requiring them to train themselves and build up their own expertise. In this case, residents could, at best, be left navigating a potentially costly trial and error approach that may include unknowingly duplicating efforts of others. At worst it could result in in treatment companies taking advantage of vulnerable residents by suggesting costly interventions that are unlikely to be effective.

While individual experiences may vary, this study demonstrates the extraordinary efforts residents took to address cuprosolvency issues at considerable personal and financial cost (Table 5.2). The two residents spent thousands of dollars ($3,800 to $30,700) and an estimated 4 months to over 1 year of full time (40 hours/week) effort investigating and attempting to address these cuprosolvency problems. But they described the stress and mental/emotional toll as being the most burdensome part of this experience. Some of this enormous burden could have been minimized if local authorities had intervened, for example, by helping Resident B identify and address the observed variable incoming pH and resulting cuprosolvency issues.

Table 5.2: **Resident Costs and Implications.**
*Values were estimated by community partners and may vary based on factors specific to each individual. Residents already had point of use reverse osmosis (RO) systems, but additional costs would apply for residents choosing to install such systems.

		Resident A	Estimated Cost	Resident B	Estimated Cost
Interventions	Point of Use	Reverse osmosis, Bottled water	$2,000	Reverse osmosis, Bottled water	$800
	Whole House Implemented	Grounding rod, Dielectric union, Sacrificial anode	$6,000	Soda ash feed to increase pH, Reagents	$1,200
	Whole House Considered	PVC insulation for copper pipe, Pipe replacement with PVC	$20,000	Ion Exchange, GAC Treatment	$1,000 to $3,000
Additional Costs	Testing	Meter to measure current, Private laboratory tests, Field colorimeter	$700	Private laboratory tests, Field colorimeter	$800
	Veterinary Bills	Diagnosis and treatments	$22,000	Diagnosis and treatments	$1,000
Other Impacts	Time	>1 year of dedicated time		~ 4 months of dedicated time	
	Stress	Major toll on mental health due to so much effort leading to no changes and feeling like no one cared or would help. Felt horrible, gaslit, stonewalled, threatened, betrayed, ignored, helpless, hopeless, and like this experience took part of their soul.		Huge amount	

Although our study developed guidance to help residents address these concerns, we also demonstrated that determining appropriate interventions may need to be on a case-by-case or community specific basis and may take substantial effort. For example, conventional flushing (up to 2 minutes) was inadequate to minimize exposure from service lines (lead),[172] and this study demonstrates that even up to 5 minutes of flushing was not enough to reduce copper below the AL in one home. Home plumbing configurations and residential concerns about water use, particularly in water stressed regions, need to be considered to develop appropriate flushing guidance. These experiences highlight the need for additional guidance, programs, and resources to reduce the burden on residents and help them determine the most effective interventions. Ideally local water authorities would serve, at least, as a clearinghouse, compiling experiences specific to their water and guiding residents toward effective

treatment techniques or away from those which were a waste of money.

Finally, this study illustrated several challenges in developing and implementing guidance. We had hoped that short term water treatments would be effective at facilitating the formation of a protective scale, as was reported in lab scale testing for granular activated carbon removal of natural organic matter.[44] However, our results demonstrate that long-term interventions (>36 weeks) may be necessary to control cuprosolvency for all treatments tested. Indeed, some corrosion control treatments, such as orthophosphate addition, must be continued indefinitely to maintain the CCT benefit, putting an ongoing burden on residents to maintain the system and continue providing reagents.[44, 302] We also demonstrated additional considerations such as fluctuations in incoming water quality, difficulty maintaining target water quality conditions in at-home systems, and the unexpected presence of polyphosphate anti-scalant additives in reagents, which could create other advantages and disadvantages for consumers.[203] Further research is needed to develop easier at-home treatment strategies and monitor their effectiveness.

Acknowledgements:

We are grateful for our community partners for their invaluable assistance in sampling and trying interventions in their homes and performing at-home tests of their water quality. Further, we are thankful to their local water utility for sending test water for laboratory studies.

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Chapter 6

Engineering Significance and Conclusions

6.1 Engineering Significance

This technical content in Chapters 2-5 of this work yield the following key intellectual contributions:

1. This work developed a protocol for systematically testing the accuracy of in-situ or at-home tests to detect dissolved and particulate lead in drinking water and proposed new methods to improve detection of particulate lead. The approach can serve as a basis to vet new tests in terms of false positives, false negatives, and poor recoveries and to potentially certify accuracy of these tests, analogous to the NSF/ANSI 53 certification for water filter lead removal (Chapter 2).[237]

2. Although particulate lead release in drinking water is sporadic, it may represent a much greater lead exposure, and therefore greater health threat, than dissolved lead, but it is also difficult to detect.[335] A novel method was developed using at-home acids to facilitate detection of particulate lead using binary at-home test kits. This may improve the detection of particulate lead, but still cannot detect lead particles that are resistant to weak acid dissolution.

3. This work critically evaluated an approach proposed by the 2015 National Drinking Water Advisory Council Consensus statement, to delineate waters as aggressive or non-aggressive to new copper pipes.[15] Water quality criteria likely to reduce cuprosolvency based on achieving a "minimum pH" or "minimum orthophosphate" dose as a function of alkalinity were developed based on extensive copper tube cuprosolvency tests. Such an approach has merit as a utility "best practice" or to assist consumers in dealing with individual buildings that have high cuprosolvency.

4. Orthophosphate corrosion control was demonstrated to be effective at quickly reducing cuprosolvency and reaching a pseudo steady state. Therefore the "minimum" orthophosphate approach is generally expected to yield a response within weeks of implementing orthophosphate treatment. In comparison, results demonstrated that it may take years or even decades to reach pseudo stead state with the "minimum" pH approach.

5. This work demonstrates the importance of additional factors that affect cuprosolvency when using the "minimum pH" approach. For example, exceptional cases were demonstrated where higher cuprosolvency was observed with higher pH. This demonstrates that further study is needed to account for interdependencies of a variety of potentially minor factors in addition to the effects of recognized key parameters (orthophosphate, alkalinity, and pH).

6. Residential guidance was developed to help assess and address cuprosolvency issues on a single-building basis. Case studies demonstrate how consumers could use this framework to address cuprosolvency issues on their own. Such an approach is highly inefficient, when compared to outreach efforts that could be coordinated by the utility, who could synthesize experiences about what works and does not work in a particular water and who has access to expert knowledge about the variability of water chemistry

across their distribution system. Such guidance is also needed for private well owners, who must address problems either on their own, or with the assistance and expertise of outreach programs (i.e. health departments or university cooperative extensions).

6.2 Conclusions

1. Most at-home lead in water test kits could not reliably detect levels of lead commonly found in drinking water (i.e. near the 15 ppb lead action level).

2. Binary kits identifying a water as either above or below a given threshold, were the most effective at-home test for residential detection of dissolved lead above the EPA lead action level in drinking water. When paired with at-home acid dissolution, binary kits may also be helpful in detecting lead from particles. These kits accurately indicated lead above the AL 78% of the time when considering total (dissolved + particulate) lead and 93% of the time when considering only dissolved lead.

3. Linear regressions representing the "minimum" pH and orthophosphate doses that constitute effective cuprosolvency control in new copper pipe (worst case values within 4-35 weeks of pipe aging) were estimated as a function of alkalinity (R^2>0.98). The "minimum" orthophosphate regression is generally in agreement with bin criteria suggested by the 2015 National Drinking Water Advisory Council Consensus Statement, but sometimes predicted that higher orthophosphate doses would be needed to control cuprosolvency. The "minimum pH" regression almost always predicted higher pH values needed than those proposed by the advisory council, which is partly due to success in formulating a "worst case" water for use in the laboratory testing.[15] Such criteria might serve as the basis for voluntary guidance to help utilities address systemwide cuprosolvency issues.

4. Orthophosphate use was very effective at reducing cuprosolvency in the short-term. However, as predicted, in some cases, higher long-term cuprosolvency was observed in orthophosphate treated waters than waters without orthophosphate corrosion control treatment. This short-term benefit and long-term detriment behavior is consistent with formation of cupric phosphate scales which are expected to have an intermediate solubility between the cupric hydroxide thought to be initially present and the more stable tenorite and malachite scales expected to form in the absence of orthophosphate.[44, 106, 114, 116, 137, 155, 179, 185, 204, 299, 301, 303, 304, 339]

5. A variety of additional factors could complicate broad application of the "minimum" pH corrosion control criteria. Consideration of the potential for calcite precipitation and pipe aging time should be accounted for. Further, results demonstrate that additional water quality parameters such as sulfate, silica, and natural organic matter may affect cuprosolvency. The interdependences of both key parameters and these additional factors need to be understood, especially since some cases were observed where higher pH yielded higher cuprosolvency, indicating that applying conventional pH corrosion control wisdom might sometimes exacerbate cuprosolvency problems.

6. Differences in performance were observed based on pipe manufacturer during development of both "minimum" pH and orthophosphate criteria, with tube from one manufacturer often yielding significantly lower cuprosolvency than tube from the other three manufacturers. Caution should be taken when utilizing copper tube cuprosolvency tests to ensure results are representative of the various types of tube present in the distribution system.

7. Cuprosolvency guidance from experiences with utilities was adapted to meet residential needs. Guidance was effectively applied in two case studies to demonstrate: 1. Use of at-home cuprosolvency tests to rule out factors other than water quality as the cause

of cuprosolvency issues and 2. Use of a whole house pH adjustment system. These case studies also highlight substantial costs and challenges residents face addressing cuprosolvency issues on their own and the potential beneficial role local authorities (e.g. utility, health department, and/or cooperative extension programs) could play in providing water source specific guidance for treatment options.

8. At-home tests could be effective for indicating elevated copper in drinking water and measuring water pH to aid with determination of residential treatment strategies. The field colorimeter is recommended for copper measurement because it had the highest accuracy of the at-home tests for copper in drinking water and exhibited relatively low variability and no false negative results in comparison to the 1.3 mg/L action level. Liquid and color tile copper tests were slightly less accurate but exhibited greater variability. The liquid pH test was also relatively accurate.

9. It has been hypothesized that short-term single building treatments could be effective at facilitating the formation of a permanent, low cuprosolvency, protective scale. However, results indicate that pH adjustment, granular active carbon, anion exchange, and reverse osmosis treatments were not effective at forming permanent protective scales for the water quality tested. Long-term and maybe permanent treatment (>36 weeks) would be required to maintain reduced cuprosolvency reductions.

6.3 Future Work

Future work is needed to address a variety of gaps in the US EPA Lead and Copper Rule. Although recent revisions increased lead sampling to include childcare facilities and schools, the LCR still leaves many residents vulnerable to elevated lead and copper in their drinking water.[27] Additional development and application of sampling strategies for lead and copper

could help reveal the true extent of corrosion issues that may currently be overlooked by current LCR sampling and estimate potential residential exposures to elevated lead and copper.

This work indicated that at-home lead and copper test kits could sometimes help residents take responsibility for mitigating their health risks. However, there is currently no certification authority or accepted test method to ensure accuracy of these test kits. Further development of these at-home test kits is needed to facilitate detection of drinking water relevant concentrations and forms (e.g. dissolved vs. particle) of lead and copper and address potential interferences.

This work developed guidance to help proactive utilities and residents address issues with elevated cuprosolvency in drinking water based on key water quality criteria. Additional research is needed to determine appropriate applications of various types of cuprosolvency test and to address challenges observed using a fresh-solids approach. Further, work identifying solids formed, especially X-ray amorphous solids, as well as determination of reaction mechanisms and thermodynamic data would aid with deeper scientific understanding and modeling of scale formation and aging in copper drinking water pipes. Results indicate that, in addition to key parameters (pH, alkalinity, orthophosphate), factors such as polyphosphate, natural organic matter, sulfate, silica, aging time, and tube manufacturer affect cuprosolvency. More research is needed to address the potential effects and interdependencies of these water quality parameters on cuprosolvency to render this guidance more broadly applicable. Finally, additional research is needed to determine the efficacy treatment strategies, including relatively untested interventions like granular activated carbon or anion exchange filtration, to address cuprosolvency issues on a residential/single building basis.

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Appendices

Appendix A

Supplemental Information for Chapter 2

A.1 User confidence in reading at-home measurements

Participants recorded results using the 16 brands of test kits for the case of relatively high dissolved lead in phase 1 along with their confidence in their measurement. Confidence values ranged from a score of one ("Not at all") to five ("Extremely"). Confidence was generally lower for the color-based tests, which required matching the sample or tile color to a color-gradient scale ranging from light yellow to red. The vial test (one brand) and one of the color strips had median confidence scores of 1 (n=6 participants), while the other color strips (10 brands) had median confidence scores of 2.5-3 (n=5-6 participants). The large intervals between measurements (range of 0 to 500 mg/L with increments of 20-300 mg/L) may have lead to potential reading errors. Binary tests had the highest confidence ratings, likely because lead presence was indicated either by a distinct binary color change from white to red or the appearance of indicator lines. The binary tests (three brands) had median confidence ratings of 3-3.5 and the binary color test (one brand) had a median rating of 4 (n=5-6 participants).

Table A.1: **User confidence in reading at-home measurements.** User confidence was reported from 5-6 participants for each test kit. Test kits include the vial test (V), 11 color strips (C1-C11), the binary color test (BC) and three binary strips (B1-B3).

Test Kit	Min	Median	Max
B1	1	3	4
B2	2	3.5	5
B3	3	3.5	4
V	1	1	4
BC	2	4	5
C1	1	1	4
C2	1	2.5	3
C3	2	3	3
C4	1	3	3
C5	2	3	3
C6	2	3	4
C7	1	3	4
C8	1	3	4
C9	2	3	4
C10	2	3	4
C11	2	3	4

A.2 Characterization of particle lead content

Three to four samples were used to determine the variability in particle composition for particles used in Phases 3 and 4. The total recoverable particle mass was defined as the sum of the masses of eight metals and phosphorous (Table A.3) in a fully digested sample. Because phosphorous could be present in particles, but was also added in the synthetic drinking water, the total recoverable particle mass was calculated using the difference between sample phosphorous concentrations and the initial dissolved phosphorous concentrations (earliest filtered phosphorous value) to differentiate phosphorous in particles from dissolved phosphorous added in the water. The percentage of lead in the total recovered particle mass was calculated as the total lead divided by the total recovered particle mass. A summary of the data is presented in Table A.2, with more detailed particle analysis in Table A.3.

Table A.2: **Leaded particle sources and characterization of lead content.** Pb: lead; Al: aluminum; Fe: iron; Zn: zinc; Sn: tin; PO_4: phosphate; LSL: Lead Service Line. The % of total recovered particle mass could not be calculated for Fe-PO_4 particles because they were collected in water, so the total particle mass added is unknown.

Particle Type	Source Type	Source Location	Mean % Total Recovered Particle Mass (SD)	% Lead of Total Recovered Particle Mass		
				Min	Mean (SD)	Max
Pb	Lead pipe	Laboratory	93% (3%)	92%	97% (4%)	103%
Al-Pb-1	LSL	Buffalo, NY	39% (4%)	23%	26% (3%)	30%
Zn-Fe	Galvanized iron pipe	Flint, MI	29% (6%)	0.8%	1.2% (0.3%)	1.4%
Pb-Sn	Lead solder	Flint, MI	47% (18%)	42%	58% (14%)	70%
Al-Pb-2	LSL	Flint, MI	34% (9%)	9%	19% (9%)	27%
Pb-Fe	LSL	Washington, D.C.	69% (9%)	55%	62% (6%)	67%
Fe-PO_4	Particles in water	Cicero, IL	-	3%	9% (6%)	14%

Table A.3: **Metals and phosphorous analysis for samples and blanks for particulate testing in phases 3 and 4.**
Pb: lead; Al: aluminum; Fe: iron; Mn: manganese; Ni: nickel; Cu: copper; Zn: zinc; Sn: tin; P: phosphorous; PO_4: phosphate
– denotes values not calculated because no particles were added to these samples (blank or particles collected in water)
* Calculated using the difference between sample phosphorous concentration and initial dissolved phosphorous (earliest filtered phosphorous value) to differentiate phosphorous in particle form from dissolved phosphorous added in water.

Phase	Particle type	Round	Sample treatment	Pb (ppb)	Al (ppb)	Fe (ppb)	Mn (ppb)	Ni (ppb)	Cu (ppb)	Zn (ppb)	Sn (ppb)	P (ppb)	Total Particle Mass Added (mg/L)	Total recovered particle mass (mg/L)*	% Recovered
Phase 3: Particulate Lead	Blank	1	Dissolved	0.4	<10.0	1,609.4	22.0	4.7	40.7	9.1	0.1	973.2	-	-	-
			Total Recovered	0.6	<10.0	8.1	0.6	0.5	33.6	8.9	0.1	983.8	-	-	-
	Pb	1	Dissolved 1 hour	1.5	<10.0	<5.0	0.1	0.4	10.2	2.3	<0.1	1,065.5	5.8	0.0	0%
			Dissolved 24 hour	4.7	10.8	<5.0	0.1	0.4	9.7	2.3	<0.1	1,028.8		0.0	0%
			Total Recovered	5,231.8	11.5	18.5	0.2	0.4	18.4	4.6	0.2	1,223.9		5.4	94%
		2	Dissolved	1.3	<10.0	<5.0	0.5	0.5	32.7	9.0	<0.1	1,004.0	4.9	0.0	1%
			Total Recovered	4,391.5	22.9	25.5	0.6	0.5	44.2	9.7	0.3	1,030.6		4.5	92%
	Al-Pb-1	1	Dissolved 1 hour	0.4	<10.0	16.9	0.3	0.4	8.5	1.6	<0.1	1,024.8	5.6	0.0	1%
			Dissolved 24 hour	1.1	12.7	<5.0	0.1	0.4	7.7	2.1	<0.1	1,043.9		0.0	1%
			Total Recovered	508.0	1,025.5	174.7	22.0	1.4	13.4	3.3	0.2	1,456.1		2.2	39%
		2	Dissolved	0.5	<10.0	11.9	0.6	0.5	30.5	9.0	<0.1	974.9	5.2	0.0	1%
			Total Recovered	495.9	790.6	177.2	20.7	1.9	36.0	8.5	0.3	1,227.9		1.8	34%
	Zn-Fe	1	Dissolved 1 hour	0.5	<10.0	8.4	0.2	0.4	9.4	4.9	<0.1	1,033.9	5.2	0.0	1%
			Dissolved 24 hour	0.3	11.6	<5.0	0.1	0.3	8.6	7.2	<0.1	1,033.6		0,0	1%
			Total Recovered	22.6	300.7	327.3	3.7	0.7	16.6	845.4	0.2	1,266.7		1.8	34%
		2	Dissolved	0.4	<10.0	44.1	0.9	0.6	32.3	10.5	<0.1	1,009.9	5.3	0.1	2%
			Total Recovered	15.0	249.0	269.1	4.2	0.8	38.8	622.8	0.2	1,059.9		1.3	24%
	Fe-PO4	1	Dissolved	2.8	21.7	<5.0	0.4	2.3	5.0	6.7	0.3	305.5	-	-	-
			Total Recovered	37.3	79.5	120.2	3.1	4.8	19.8	17.6	1.3	376.3		-	-
	Blank	2	Dissolved <30 min	0.5	<10.0	<5.0	0.1	0.4	8.7	2.1	<0.1	1,002.7	-	-	-
			Dissolved 1 hour	0.3	<10.0	<5.0	0.1	0.4	9.8	2.6	<0.1	1,054.5		-	-
			Dissolved 24 hour	0.2	11.1	<5.0	0.1	0.4	8.5	2.2	<0.1	1,053.9		-	-
			Total Recovered	0.4	<10.0	17.7	0.2	0.5	12.6	2.3	0.1	1,186.9		-	-
	Pb-Sn	1	Dissolved <30 min	33.7	<10.0	<5.0	<0.1	0.5	4.5	<1.0	0.1	912.6	5.2	0.0	1%
			Dissolved 1 hour	27.4	<10.0	<5.0	<0.1	0.5	4.5	<1.0	<0.1	882.4		0.0	0%
			Dissolved 24 hour	9.7	11.7	<5.0	<0.1	0.5	6.7	<1.0	0.2	819.7		0.0	-1%
			Total Recovered	2,041.8	<10.0	30.9	0.3	1.0	188.8	17.9	480.2	1,078.9		2.9	56%
	Al-Pb-2	1	Dissolved <30 min	0.4	<10.0	<5.0	0.1	0.4	8.8	2.1	<0.1	1,086.6	5.2	0.0	0%

Prior dissolution steps are not expected to affect particle characterization results since samples were analyzed after full digestion using 2% nitric acid and 2% hydroxylamine and heated for at least 1 week. Therefore, samples with and without dissolution steps (lemon juice or vinegar) prior to full digestion were included in the analysis. During testing, kits from one brand of binary kit were expired when received, and were not used. Once replacements for these kits were received, a second round of experiments were performed to test these binary strips with the three particle types that had been previously tested with the other two brands of binary strip.

Variability in the percentage of lead in the total recovered particle mass was observed among the different particles, with average lead content ranging from 0.8% to 92% (Table A.2). However, variability among replicates (particles from the same source) exhibited much less variability, with standard deviations ranging from 0.3% to 14%.

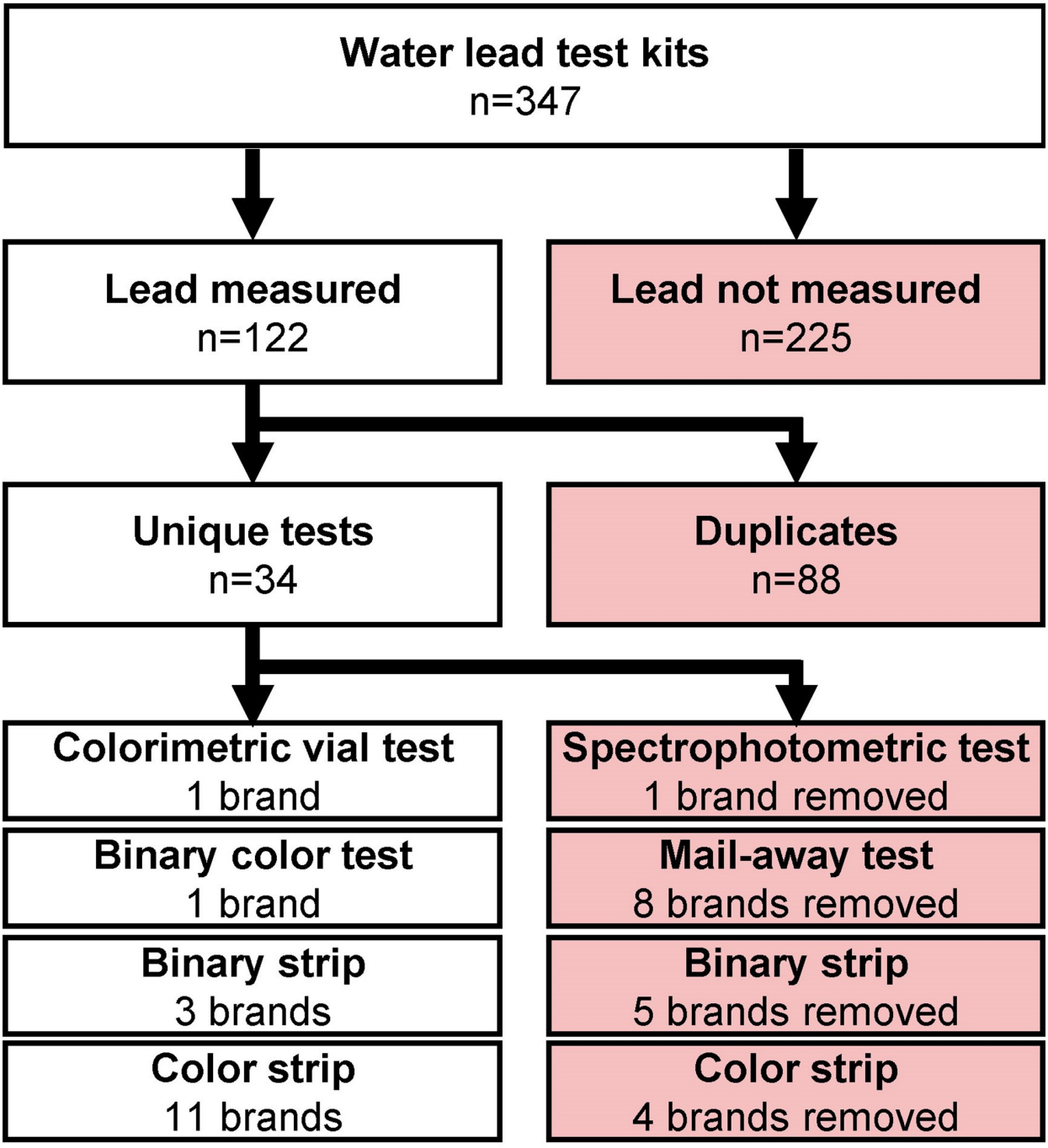


Figure A.1: **Selection of test kits for analysis.** Red boxes indicate test kits that were removed.

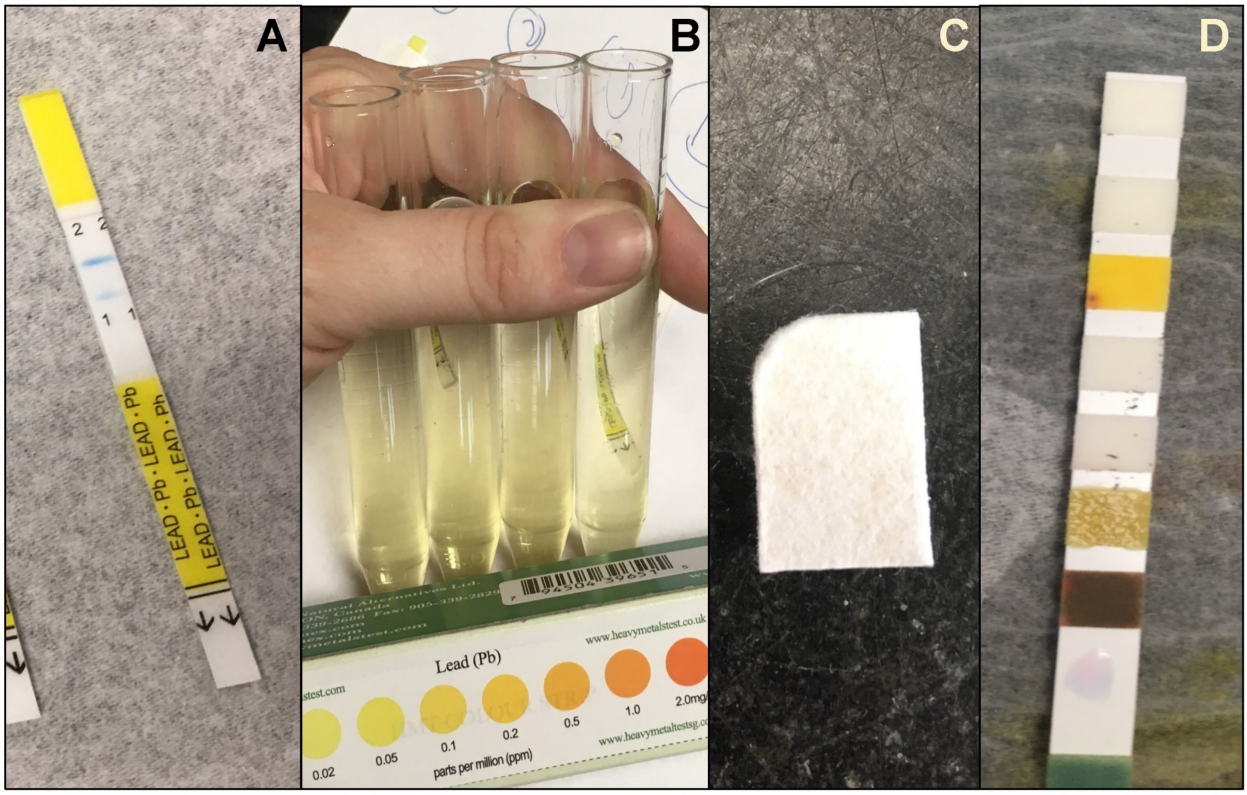


Figure A.2: **Example of the test kits analyzed.** (A) binary strip, (B) colorimetric vial test, (C) binary color test, and (D) color strip.

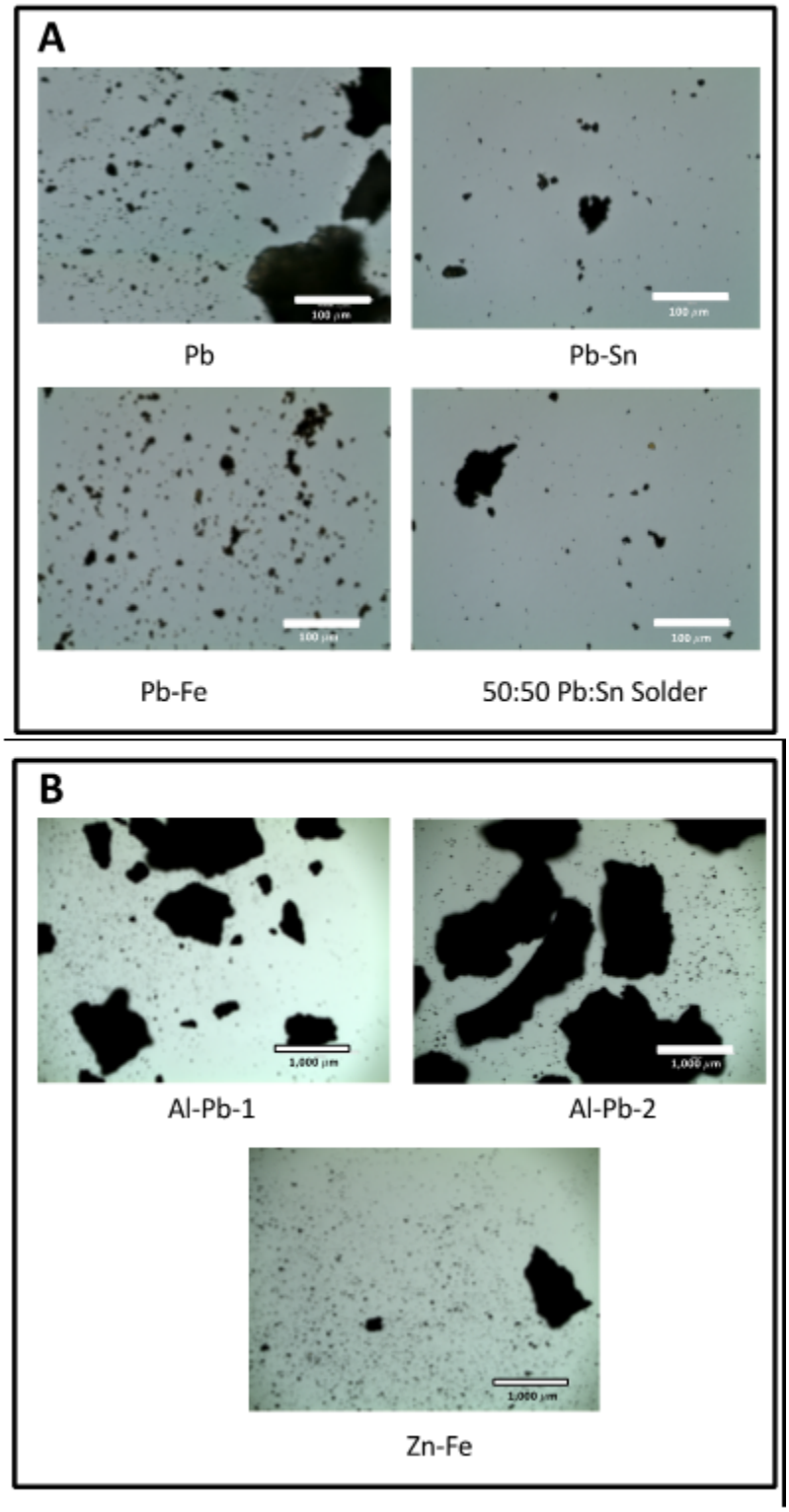


Figure A.3: **Representative images of particles determined through raman atomic force microscopy.** Images are presented at a A) 40x and B) 4x magnifications, depending on particle size. Pb: lead; Sn: Tin; Fe: iron; Al: aluminum; Zn: zinc.

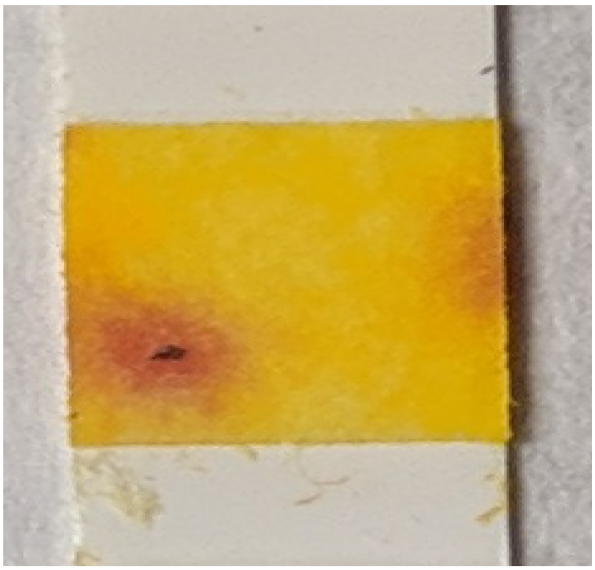


Figure A.4: **Color change on the tile of a color strip when in contact with a lead solder particle**

Phase	Particle type	Round	Sample treatment	Pb (ppb)	Al (ppb)	Fe (ppb)	Mn (ppb)	Ni (ppb)	Cu (ppb)	Zn (ppb)	Sn (ppb)	P (ppb)	Total Particle Mass Added (mg/L)	Total recovered particle mass (mg/L)*	% Recovered
Phase 3: Particulate Lead	Blank	1	Dissolved	0.4	<10.0	1,609.4	22.0	4.7	40.7	9.1	0.1	973.2	-	-	-
			Total Recovered	0.6	<10.0	8.1	0.6	0.5	33.6	8.9	0.1	983.8	-	-	-
	Pb	1	Dissolved 1 hour	1.5	<10.0	<5.0	0.1	0.4	10.2	2.3	<0.1	1,065.5	5.8	0.0	0%
			Dissolved 24 hour	4.7	10.8	<5.0	0.1	0.4	9.7	2.3	<0.1	1,028.8		0.0	0%
			Total Recovered	5,231.8	11.5	18.5	0.2	0.4	18.4	4.6	0.2	1,223.9		5.4	94%
		2	Dissolved	1.3	<10.0	<5.0	0.5	0.5	32.7	9.0	<0.1	1,004.0	4.9	0.0	1%
			Total Recovered	4,391.5	22.9	25.5	0.6	0.5	44.2	9.7	0.3	1,030.6		4.5	92%
	Al-Pb-1	1	Dissolved 1 hour	0.4	<10.0	16.9	0.3	0.4	8.5	1.6	<0.1	1,024.8	5.6	0.0	1%
			Dissolved 24 hour	1.1	12.7	<5.0	0.1	0.4	7.7	2.1	<0.1	1,043.9		0.0	1%
			Total Recovered	508.0	1,025.5	174.7	22.0	1.4	13.4	3.3	0.2	1,456.1		2.2	39%
		2	Dissolved	0.5	<10.0	11.9	0.6	0.5	30.5	9.0	<0.1	974.9	5.2	0.0	1%
			Total Recovered	495.9	790.6	177.2	20.7	1.9	36.0	8.5	0.3	1,227.9		1.8	34%
	Zn-Fe	1	Dissolved 1 hour	0.5	<10.0	8.4	0.2	0.4	9.4	4.9	<0.1	1,033.9	5.2	0.0	1%
			Dissolved 24 hour	0.3	11.6	<5.0	0.1	0.3	8.6	7.2	<0.1	1,033.6		0,0	1%
			Total Recovered	22.6	300.7	327.3	3.7	0.7	16.6	845.4	0.2	1,266.7		1.8	34%
		2	Dissolved	0.4	<10.0	44.1	0.9	0.6	32.3	10.5	<0.1	1,009.9	5.3	0.1	2%
			Total Recovered	15.0	249.0	269.1	4.2	0.8	38.8	622.8	0.2	1,059.9		1.3	24%
	Fe-PO4	1	Dissolved	2.8	21.7	<5.0	0.4	2.3	5.0	6.7	0.3	305.5	-	-	-
			Total Recovered	37.3	79.5	120.2	3.1	4.8	19.8	17.6	1.3	376.3		-	-
	Blank	2	Dissolved <30 min	0.5	<10.0	<5.0	0.1	0.4	8.7	2.1	<0.1	1,002.7	-	-	-
			Dissolved 1 hour	0.3	<10.0	<5.0	0.1	0.4	9.8	2.6	<0.1	1,054.5		-	-
			Dissolved 24 hour	0.2	11.1	<5.0	0.1	0.4	8.5	2.2	<0.1	1,053.9		-	-
			Total Recovered	0.4	<10.0	17.7	0.2	0.5	12.6	2.3	0.1	1,186.9		-	-
	Pb-Sn	1	Dissolved <30 min	33.7	<10.0	<5.0	<0.1	0.5	4.5	<1.0	0.1	912.6	5.2	0.0	1%
			Dissolved 1 hour	27.4	<10.0	<5.0	<0.1	0.5	4.5	<1.0	<0.1	882.4		0.0	0%
			Dissolved 24 hour	9.7	11.7	<5.0	<0.1	0.5	6.7	<1.0	0.2	819.7		0.0	-1%
			Total Recovered	2,041.8	<10.0	30.9	0.3	1.0	188.8	17.9	480.2	1,078.9		2.9	56%
	Al-Pb-2	1	Dissolved <30 min	0.4	<10.0	<5.0	0.1	0.4	8.8	2.1	<0.1	1,086.6	5.2	0.0	0%

Phase 4: Lemon Juice Dissolution																
			Dissolved 1 hour	0.5	<10.0	<5.0	<0.1	0.4	8.7	1.8	<0.1	1,065.0		0.0	0%
			Dissolved 24 hour	1.3	12.0	<5.0	0.1	0.4	8.0	1.9	<0.1	1,041.6		0.0	0%
			Total Recovered	292.0	644.0	107.9	13.9	3.0	42.1	24.6	0.7	1,362.1		1.4	27%
	Pb-Fe	1	Dissolved <30 min	15.5	<10.0	<5.0	0.1	0.5	8.5	2.0	<0.1	1,055.2		0.0	1%
			Dissolved 1 hour	19.4	<10.0	<5.0	0.1	0.5	8.6	2.0	<0.1	1,079.0	5.4	0.1	1%
			Dissolved 24 hour	25.6	15.5	<5.0	<0.1	0.5	7.3	1.5	<0.1	1,015.5		0.0	0%
			Total Recovered	2,302.5	185.1	475.0	47.5	1.3	111.1	19.0	9.8	1,315.4		3.4	63%
	Blank	1	Dissolved 1 hour	0.5	<10.0	28.2	7.4	1.0	38.8	32.9	<0.1	4,076.5	-	-	-
			Dissolved 24 hour	1.3	<10.0	34.7	7.6	1.1	40.2	39.6	0.5	4,013.2		-	-
			Total Recovered	0.6	<10.0	32.4	7.1	1.0	40.6	23.5	0.1	4,129.2		-	-
	Pb	1	Dissolved 1 hour	608.2	12.6	30.8	7.7	1.7	42.3	193.0	0.2	4,051.7	5.0	0.9	18%
			Dissolved 24 hour	2,782.3	12.9	34.2	7.6	1.1	45.8	39.3	0.4	4,058.1		2.9	59%
			Total Recovered	4,466.9	<10.0	37.3	7.0	1.0	45.4	26.4	<0.1	4,294.2		4.8	97%
	Al-Pb-1	1	Dissolved 1 hour	70.0	<10.0	31.1	8.2	1.1	40.9	42.5	<0.1	4,262.9	4.8	0.2	4%
			Dissolved 24 hour	385.2	235.9	46.8	18.1	1.2	43.8	39.2	0.5	4,130.3		0.6	13%
			Total Recovered	501.1	869.2	196.7	30.8	1.9	40.8	23.5	<0.1	4,684.0		2.1	43%
	Zn-Fe	1	Dissolved 1 hour	3.0	36.6	40.4	7.6	1.0	41.5	201.7	0.2	3,815.7	5.2	0.3	6%
			Dissolved 24 hour	13.0	235.0	153.3	9.8	1.4	50.0	650.3	0.4	3,946.0		1.2	24%
			Total Recovered	14.4	241.6	260.3	9.7	1.4	44.2	651.5	<0.1	4,431.3		1.8	35%
	Fe-PO4	1	Dissolved 1 hour	34.0	48.7	77.7	11.6	2.1	28.6	53.8	0.7	4,352.3	-	-	-
			Dissolved 24 hour	35.4	57.7	108.5	12.1	2.2	38.8	56.6	2.8	4,316.7		-	-
			Total Recovered	35.7	89.4	164.7	12.2	2.4	31.3	51.9	2.5	5,278.2		-	-
	Pb-Sn	1	Dissolved 1 hour	1,460.2	<10.0	30.1	6.7	1.3	86.4	35.3	72.2	4,526.5	5.0	1.7	34%
			Dissolved 24 hour	1,698.9	13.4	24.3	6.7	1.5	109.1	38.6	188.3	4,252.6		1.8	36%
			Total Recovered	1,781.3	14.3	91.7	8.2	1.8	204.9	34.0	166.3	5,143.0		2.9	58%
	Al-Pb-2	1	Dissolved 1 hour	36.5	22.9	32.6	7.6	1.0	21.7	26.9	0.4	4,385.8	5.4	0.1	3%
			Dissolved 24 hour	172.9	331.1	63.4	12.3	2.4	41.1	33.2	0.8	4,313.3		0.6	11%
			Total Recovered	228.1	680.9	274.4	20.3	3.4	50.9	36.2	1.2	5,517.9		2.4	45%
	Pb-Fe	1	Dissolved 1 hour	869.6	50.6	67.2	19.7	1.1	70.1	28.8	1.2	4,430.1	5.2	1.1	21%
			Dissolved 24 hour	1,770.2	117.8	260.6	39.7	1.3	99.5	29.9	1.6	4,656.8		2.5	49%
			Total Recovered	2,278.2	153.3	412.6	46.1	1.5	114.3	26.3	2.8	5,511.3		4.1	79%
Figure 4: Sugar Solution	Blank	1	Dissolved 1 hour	0.6	<10.0	<5.0	0.9	0.9	33.6	19.3	<0.1	2,873.0	-	-	-

			Dissolved 24 hour	1.5	<10.0	7.5	0.9	0.9	48.0	24.5	0.4	2,688.2		-	-
			Total Recovered	0.6	13.0	<5.0	0.8	0.8	32.1	12.0	0.1	2,656.4		-	-
	Pb	1	Dissolved 1 hour	435.1	<10.0	12.2	1.0	1.1	34.2	25.8	<0.1	2,748.7	5.2	0.5	10%
			Dissolved 24 hour	1,111.3	<10.0	<5.0	0.9	0.9	32.7	22.9	0.4	2,571.5		1.0	19%
			Total Recovered	4,733.4	<10.0	52.4	1.4	0.9	42.5	16.5	6.3	2,488.4		4.6	88%
	Al-Pb-1	1	Dissolved 1 hour	73.4	<10.0	471.2	5.2	1.5	34.2	23.5	0.1	2,666.8	5.0	0.6	12%
			Dissolved 24 hour	308.0	44.1	8.3	1.4	0.9	32.5	21.1	0.3	2,454.8		0.2	4%
			Total Recovered	591.0	979.3	199.7	28.5	1.8	31.7	10.4	0.5	2,769.0		1.9	39%
	Zn-Fe	1	Dissolved 1 hour	0.9	<10.0	5.6	0.9	0.8	30.4	112.0	<0.1	2,639.9	5.0	0.2	3%
			Dissolved 24 hour	4.9	48.4	7.0	1.4	1.0	33.5	491.7	0.6	2,762.5		0.7	14%
			Total Recovered	17.1	222.8	338.1	3.1	1.8	35.5	671.6	0.3	2,608.4		1.3	25%
	Fe-PO4	1	Dissolved 1 hour	31.7	49.7	28.4	2.1	2.3	14.2	25.3	0.5	2,790.1	-	-	-
			Dissolved 24 hour	31.4	54.8	46.0	2.7	1.6	15.6	34.3	0.7	2,599.6		-	-
			Total Recovered	34.4	67.8	279.1	5.1	2.0	17.8	29.1	1.1	2,600.3		-	-
	Pb-Sn	1	Dissolved 1 hour	423.8	<10.0	<5.0	0.7	0.6	48.2	13.7	0.6	2,895.9	5.2	0.5	10%
			Dissolved 24 hour	493.8	14.0	31.7	1.1	0.6	65.0	11.0	2.4	3,033.4		0.8	15%
			Total Recovered	598.9	<10.0	25.7	0.8	0.7	77.2	7.6	186.1	3,400.1		1.4	27%
	Al-Pb-2	1	Dissolved 1 hour	29.8	<10.0	<5.0	0.7	0.5	10.9	12.8	0.1	2,887.6	5.8	0.1	1%
			Dissolved 24 hour	201.3	43.2	<5.0	1.4	0.8	15.6	14.2	0.1	3,060.6		0.5	8%
			Total Recovered	492.6	597.3	106.1	23.9	2.9	41.8	25.2	0.6	3,396.5		1.8	31%
	Pb-Fe	1	Dissolved 1 hour	492.7	21.2	13.5	1.2	0.6	35.8	15.5	<0.1	2,980.0	4.8	0.6	12%
			Dissolved 24 hour	926.5	66.4	8.8	3.6	0.7	64.3	14.3	0.1	3,014.4		1.1	23%
			Total Recovered	1,920.4	144.3	380.1	39.0	1.1	101.0	14.0	3.7	3,413.8		3.0	63%

Table A.4: **Size distributions of leaded particles determined through raman atomic force microscopy at a 40x zoom.**
Pb: lead; Fe: iron; Sn: tin

	Pb		Pb-Sn		Pb-Fe		Lead Solder	
Size	Number of Particles	% of Total Particles	Number of Particles	% of Total Particles	Number of Particles	% of Total Particles	Number of Particles	% of Total Particles
0.5-1 μm	1593	20%	333	20%	189	22%	707	16%
1-2 μm	2488	31%	471	28%	239	28%	1140	25%
2-5 μm	2809	35%	581	35%	265	31%	1709	38%
5-10 μm	886	11%	193	12%	95	11%	720	16%
10-15 μm	211	3%	41	2%	33	4%	132	3%
15-50 μm	93	1%	38	2%	41	5%	103	2%
50+ μm	21	0%	3	0%	1	0%	40	1%
Total	8101	-	1660	-	863	-	4551	-

Table A.5: **Size distributions of leaded particles determined through raman atomic force microscopy at a 4x zoom.**
Pb: lead; Al: aluminum; Fe: iron; Zn: Zinc

	Al-Pb-1		Al-Pb-2		Zn-Fe	
Size	Number of Particles	% of Total Particles	Number of Particles	% of Total Particles	Number of Particles	% of Total Particles
5-10 μm	3526	38%	6961	53%	9913	42%
10-20 μm	3371	36%	4143	32%	8461	36%
20-40 μm	1604	17%	1813	14%	3616	15%
40-100 μm	515	6%	166	1%	1018	4%
100+ μm	262	3%	60	0%	404	2%
Total	9,278	-	13,143	-	23,412	-

Table A.6: **Metals and phosphorous analysis for samples and blanks in phases 1 and 2 testing.** Test kits include the vial test (V), 11 color strips (C1-C11), the binary color test (BC1) and three binary strips (B1-B3).
– denotes values not analyzed for that sample.
Pb: lead; Al: aluminum; Fe: iron; Mn: manganese; Ni: nickel; Cu: copper; Zn: zinc; Sn: tin; P: phosphorous
*Digestion for dissolved lead samples was for >16 hours, but for 1 week for total recovered samples

Phase	Test Kits Tested	Sample	Sample Treatment*	Pb (ppb)	Al (ppb)	Fe (ppb)	Mn (ppb)	Ni (ppb)	Cu (ppb)	Zn (ppb)	Sn (ppb)	P (ppb)
Phase 1: High Dissolved Lead	B1-B2, V, C1-C11, BC1	Blank	Dissolved	0.4	72.9	5.5	2.8	5.2	11.9	10.3	<0.1	6.2
			Total Recovered	0.7	45.0	7.0	3.0	1.3	16.3	5.9	0.4	12.1
		150 ppb dissolved Pb	Dissolved	148.8	69.8	5.6	1.8	0.6	14.9	10.0	<0.1	5.1
			Total Recovered	171.4	15.1	8.5	3.0	0.6	18.6	7.3	0.2	22.9
	B3	Blank	Dissolved	0.4	< 10.0	<5.0	2.5	0.6	23.9	8.3	<0.1	<5.0
			Total Recovered	0.5	<10.0	<5.0	2.7	0.6	30.0	7.1	0.1	<5.0
		150 ppb dissolved Pb	Dissolved	154.7	<10.0	<5.0	2.6	0.6	26.4	6.0	<0.1	<5.0
			Total Recovered	166.4	<10.0	<5.0	2.7	0.6	28.8	6.0	<0.1	151.3
Phase 2A: Low Dissolved Lead	B1-B2, V	Blank	Dissolved	0.4	<10.0	<5.0	2.5	0.3	18.8	13.3	0.1	<5.0
			Total Recovered	0.5	<10.0	<5.0	2.8	0.2	24.5	9.4	0.1	8.7
		10 ppb dissolved Pb	Dissolved	8.0	<10.0	<5.0	2.5	0.1	20.0	9.5	<0.1	<5.0
			Total Recovered	9.5	<10.0	<5.0	2.7	0.1	24.1	7.3	0.1	<5.0
		20 ppb dissolved Pb	Dissolved	16.3	<10.0	<5.0	2.5	0.2	19.7	10.8	<0.1	<5.0
			Total Recovered	18.2	223.7	<5.0	2.8	0.4	23.2	8.1	0.1	5.9
	B3	Blank	Dissolved	0.2	<10.0	6.7	1.8	0.6	13.2	4.5	<0.1	<5.0
			Total Recovered	0.4	<10.0	30.8	2.2	0.6	15.2	3.1	0.1	71.2
	B1- B3, V	5 ppb dissolved Pb	Dissolved	4.4	<10.0	21.7	2.2	0.5	14.7	3.4	<0.1	<5.0
			Total Recovered	5.0	<10.0	<5.0	2.0	0.5	16.5	4.3	0.1	76.6
	B3	10 ppb dissolved Pb	Dissolved	9.1	<10.0	145.6	19.4	0.9	16.3	3.0	<0.1	<5.0
			Total Recovered	9.8	<10.0	<5.0	23.1	0.5	16.5	2.3	<0.1	113.0
		20 ppb dissolved Pb	Dissolved	18.0	<10.0	17.7	2.9	0.6	14.4	2.7	<0.1	<5.0
			Total Recovered	19.6	<10.0	134.5	3.8	0.9	17.0	2.4	<0.1	125.5
Phase 2B: Solder Particles	C1-C11BC1	Blank	Dissolved	0.4	<10.0	<5.0	2.5	0.3	18.8	13.3	0.1	<5.0
			Total Recovered	0.5	<10.0	<5.0	2.8	0.2	24.5	9.4	0.1	8.7
		Lead Solder	Dissolved	8.6	<10.0	<5.0	2.8	0.2	18.6	9.5	0.5	<5.0
			Total Recovered	50,781.1	<10.0	146.7	3.7	1.4	28.2	9.1	NA	<5.0

Table A.7: **Characterization from one sample of treated drinking water from the Kankakee River, as received.**
$CaCO_3$: calcium carbonate; Cl: chloride; S: sulfur; Pb: lead; Al: aluminum; Fe: iron; Mn: manganese; Ni: nickel; Cu: copper; Zn: zinc; Sn: tin; P: phosphorous

Parameter	Concentration
Alkalinity (mg/L as $CaCO_3$)	50.2
Cl (ppm)	33.3
S (ppm)	70.8
Pb (ppb)	3.4
Al (ppb)	<10.0
Fe (ppb)	269.5
Mn (ppb)	2.4
Ni (ppb)	1.8
Cu (ppb)	130.9
Zn (ppb)	39.4
Sn (ppb)	1.8
P (ppb)	<5.0

Table A.8: **Lead results for semi-quantitative test kits measuring phase 1: high dissolved lead (150 ppb = 0.15 mg/L).** Test kits include the vial test (V) and 11 color strips (C1-C11). Variation in measurement is provided as standard deviation (SD).

Kit Type	Participant	Leaded Sample					Blank Sample				
		Measured Lead (mg/L)	Min (mg/L)	Mean (mg/L)	SD (mg/L)	Max (mg/L)	Measured Lead (mg/L)	Min (mg/L)	Mean (mg/L)	SD (mg/L)	Max (mg/L)
V	1	0.10 ± <0.01	0.05	0.12	0.05	0.20	0.10	0.01	0.09	0.07	0.20
	2	0.07 ± 0.03					0.01				
	3	0.10 ± <0.01					0.05				
	4	0.10 ± <0.01					0.10				
	5	0.20 ± <0.01					0.20				
	6	0.17 ± 0.06					0.05				
C1	1	20 ± <1	0	24	10	50	20	20	30	15	50
	2	20 ± <1					20				
	3	20 ± <1					20				
	4	40 ± 17					50				
	5	30 ± 17					50				
	6	13 ± 12					20				
C2	1	0 ± <1	0	2	4	10	0	0	2	4	10
	2	10 ± <1					10				
	3	0 ± <1					0				
	4	0 ± <1					0				
	5	0 ± <1					0				
	6	0 ± <1					0				
C3	1	0 ± <1	0	4	8	20	0	0	0	0	0
	2	7 ± 12					0				
	3	0 ± <1					0				
	4	0 ± <1					0				
	5	20 ± <1					0				
	6	0 ± <1					0				

C4	1 2 3 4 5 6	50 ± <1 50 ± <1 50 ± <1 20 ± <1 50 ± <1 50 ± <1	20	45	12	50	50 50 50 20 50 50	20	45	12	50
C5	1 2 3 4 5 6	0 ± <1 0 ± <1 0 ± <1 0 ± <1 0 ± <1 0 ± <1	0	0	0	0	0 0 0 0 0 0	0	0	0	0
C6	1 2 3 4 5 6	0 ± <1 0 ± <1 0 ± <1 0 ± <1 0 ± <1 0 ± <1	0	0	0	0	0 0 0 0 0 0	0	0	0	0
C7	1 2 3 4 5	0 ± <1 0 ± <1 0 ± <1 20 ± <1 13 ± <12	0	7	9	20	0 0 0 20 0	0	4	9	20
C8	1 2 3 4 5	20 ± <1 20 ± <1 0 ± <1 0 ± <1 20 ± <1	0	12	11	20	20 20 0 0 20	0	12	11	20
C9	1 2	0 ± <1 0 ± <1	0	5	9	20	0 0	0	4	9	20

	3 4 5	20 ± <1 7 ± <12 0 ± <1					20 0 0				
C10	1 2 3 4 5 6	0 ± <1 0 ± <1 0 ± <1 0 ± <1 0 ± <1 0 ± <1	0	0	0	0	0 0 0 0 0 0	0	0	0	0
C11	1 2 3 4 5 6	13 ± <12 0 ± <1 20 ± <1 0 ± <1 0 ± <1 0 ± <1	0	6	9	20	0 0 0 0 0 0	0	0	0	0

Table A.9: **Effects of CCT on particulate lead detection and dissolution.**
CCT: corrosion control treatment; Poly: Polyphosphate; Ortho: orthophosphate; Zn: zinc
- denotes no percent improvement calculated for cases with no treatment

Treatment Type	Treatment Time	CCT type	Total Lead (ppb)	Dissolved Lead (ppb)	% improved	# Positive Tests
No Treatment	**< 30 min**	Polyphosphate	243.6	**148.4 (61%)**	-	3/3
		Zn & Orthophosphate	12.0	0.4 (3%)	-	0/3
		Orthophosphate	20.1	2.5 (13%)	-	0/3
		90:10 Ortho/Poly-phosphate Blend	17.2	4.8 (28%)	-	0/3
		Polyphosphate & Nitrate	689.6	**378.0 (55%)**	-	3/3
		Unamended Control	1,347.9	**130.2 (10%)**	-	3/3
		Amended Control	706.8	**443.6 (63%)**	-	3/3
Lemon Juice Dissolution	**24 hr**	Polyphosphate	203.2	**185.5 (91%)**	30%	3/3
		Zn & Orthophosphate	2.6	2.0 (78%)	75%	0/3
		Orthophosphate	14.2	12.1 (85%)	72%	0/3
		90:10 Ortho/Poly-phosphate Blend	12.5	8.4 (67%)	39%	0/3
		Polyphosphate & Nitrate	548.9	**521.9 (95%)**	40%	2/3
		Unamended Control	425.4	**392.4 (92%)**	82%	3/3
		Amended Control	574.8	**530.3 (92%)**	29%	3/3

Table A.10: **Metals and phosphorous analysis for samples and blanks for phase 5, Kankakee, IL case study on the effects of CCT.**
CCT: corrosion control treatment; Poly: Polyphosphate; Ortho: orthophosphate; Pb: lead; Al: aluminum; Fe: iron; Mn: manganese; Ni: nickel; Cu: copper; Zn: zinc; Sn: tin; P: phosphorous
*Digestion for dissolved lead samples was for 24 hours for both no treatment and lemon juice, but for 1 week for total recovered samples

Additional Dissolution	Sample	Sample Treatment*	Pb (ppb)	Al (ppb)	Fe (ppb)	Mn (ppb)	Ni (ppb)	Cu (ppb)	Zn (ppb)	Sn (ppb)	P (ppb)
No Treatment	Unamended Control	Dissolved	130.2	21.6	<5.0	0.2	0.7	9.4	13.9	1.7	<5.0
		Total Recovered	1,347.9	20.4	11.8	0.6	1.4	65.5	100.3	3.3	10.7
	Amended Control	Dissolved	443.6	22.1	<5.0	0.2	1.0	25.3	12.9	4.6	<5.0
		Total Recovered	706.8	21.4	18.2	0.5	1.1	46.0	14.5	130.0	<5.0
	Polyphosphate	Dissolved	148.4	25.0	<5.0	0.2	0.8	26.9	73.4	1.0	477.8
		Total Recovered	243.6	35.8	13.2	2.2	0.8	54.4	79.5	1.9	510.6
	Zn & Orthophosphate	Dissolved	0.4	21.0	<5.0	0.2	0.7	56.6	451.5	0.2	915.2
		Total Recovered	12.0	32.0	6.0	0.4	0.8	96.4	512.2	1.6	980.1
	Orthophosphate	Dissolved	2.5	21.1	<5.0	0.2	0.7	71.4	123.1	0.9	931.2
		Total Recovered	20.1	22.8	6.5	0.4	0.7	126.5	135.1	4.3	978.3
	90:10 Ortho/Poly-phosphate Blend	Dissolved	4.8	21.6	<5.0	0.2	0.8	145.4	159.2	0.7	1,050.6
		Total Recovered	17.2	19.1	9.2	0.4	0.9	194.7	155.8	20.7	1,035.2
	Polyphosphate & Nitrate	Dissolved	378.0	21.2	<5.0	0.2	0.7	13.6	3.3	2.2	439.6
		Total Recovered	689.6	20.3	7.3	0.3	0.8	27.9	4.7	51.3	473.1
Lemon Juice Dissolution	Unamended Control	Dissolved	530.3	24.1	28.8	6.6	1.5	40.8	57.9	69.9	3,250.4
		Total Recovered	574.8	27.3	93.2	7.6	1.6	43.7	35.5	54.3	3,572.7
	Amended Control	Dissolved	392.4	26.0	33.9	6.7	1.4	36.4	56.3	42.5	3,634.2
		Total Recovered	425.4	33.0	50.8	7.7	1.4	39.6	50.0	48.7	3,803.0
	Polyphosphate	Dissolved	185.5	23.6	32.0	6.3	1.3	41.0	128.7	0.9	3,661.9
		Total Recovered	203.2	39.5	60.7	7.6	1.5	45.3	93.5	1.1	4,141.1
	Zn & Orthophosphate	Dissolved	2.0	24.2	35.0	6.4	1.3	76.3	488.3	1.1	4,247.5
		Total Recovered	2.6	36.1	51.8	7.3	1.3	82.9	486.2	1.2	4,610.7
	Orthophosphate	Dissolved	12.1	26.3	38.2	6.7	1.3	104.2	148.4	2.3	4,610.2
		Total Recovered	14.2	34.7	49.1	7.4	1.4	106.3	137.9	2.3	4,619.9
	90:10 Ortho/Poly-phosphate Blend	Dissolved	8.4	25.2	35.7	6.5	1.4	161.9	170.7	6.5	4,358.0
		Total Recovered	12.5	34.8	49.9	7.6	1.3	180.2	169.2	8.1	4,873.0
	Polyphosphate & Nitrate	Dissolved	521.9	26.3	34.4	6.8	1.3	27.3	31.1	18.5	3,955.8
		Total Recovered	548.9	29.5	45.1	7.6	1.4	29.4	21.9	27.1	4,288.3

Table A.11: **Contingency tables of binary strip results based on total and dissolved lead concentrations.** Analysis includes 134 binary kits tested throughout all phases during this study, excluding the 7 invalid results.

		Total Lead		Dissolved Lead	
		> 15 ppb	< 15 ppb	> 15 ppb	< 15 ppb
Binary Test Kit Result	Positive	78 (58.2%)	7 (5.2%)	78 (58.2%)	7 (5.2%)
	Negative	23 (17.2%)	26 (19.4%)	2 (1.5%)	47 (35.1%)

Appendix B

Supplemental Information for Chapter 3

B.1 Determination of "minimum" orthophosphate criteria

The minimum orthophosphate (OP) dose needed to achieve copper concentrations below the 1.3 mg/L action level were estimated based on water alkalinity. To determine this, three orthophosphate doses were tested for each of four alkalinities (n= 20 pipe segments per condition).

Results for the 90^{th}, 95^{th} and 100^{th} percentile (18^{th} 19^{th}, and 20^{th} highest) copper values after 4 and 22 weeks of tube/pipe conditioning (8 weeks with water changes and a 14-week extended stagnation period) were interpolated for each alkalinity to determine the minimum OP dose that yielded copper below the action level. An example interpolation for the 100^{th} percentile (maximum copper value) results after 22 weeks of pipe aging is presented in Figure B.1.

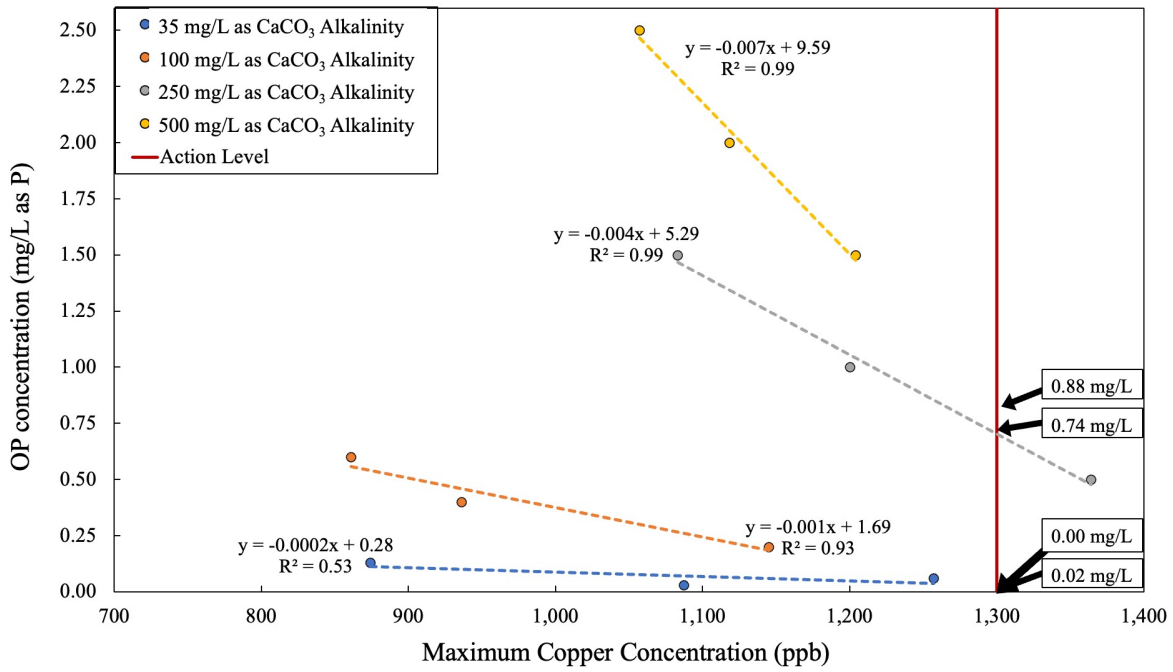


Figure B.1: **Illustrative graph for defining minimum orthophosphate dose to achieve copper concentrations below the Action Level**
for the 100th percentile (Maximum) copper concentrations after 22 weeks of pipe aging. Arrows with boxes denote the "minimum" orthophosphate (mg/L as P) predicted by the regressions to yield copper below the action level.

Linear regressions of the "minimum" OP dose needed to get copper below the action level were developed based on the water's alkalinity and "worst case" OP values. To simulate the "worst case" scenario, the higher interpolated orthophosphate value after either week 4 or week 22 was used for each alkalinity to construct the linear regressions. Regressions are provided for the 90th, 95th and 100th percentile data (Figure B.2).

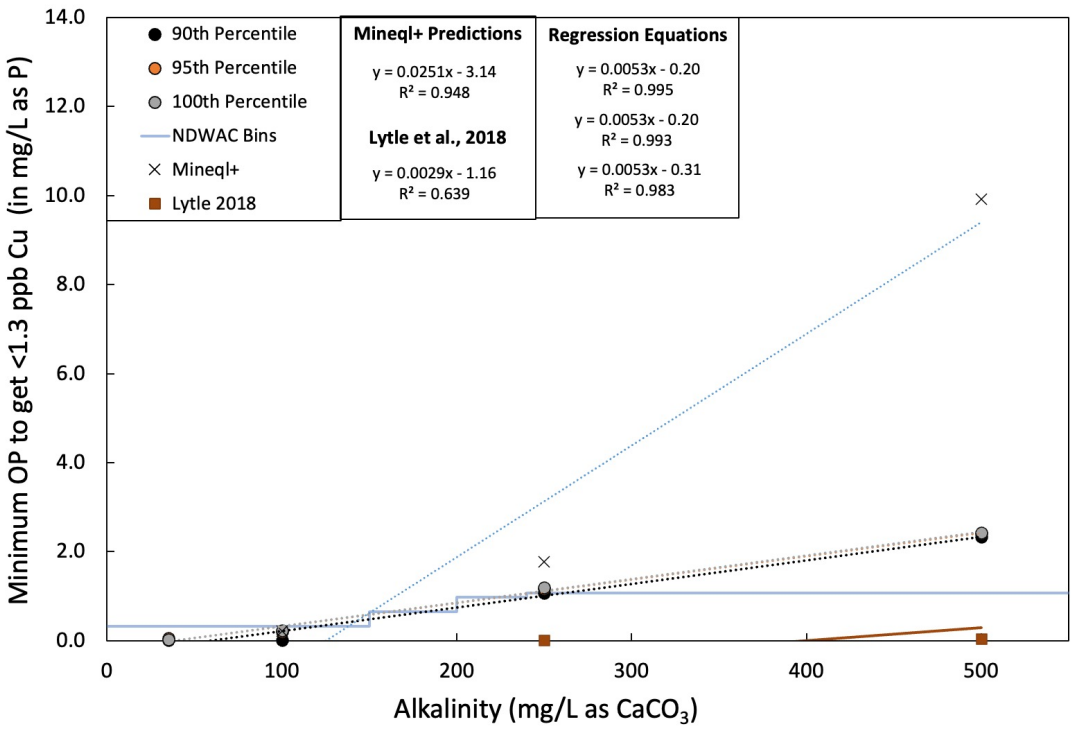


Figure B.2: **Linear regressions defining the "minimum" orthophosphate criteria** for varying alkalinities to define waters "non-aggressive" to copper. Orthophosphate values represent the highest interpolated value, after either 4 or 22 weeks of testing, from linear correlations of orthophosphate and maximum (100^{th} percentile), second (95^{th} percentile) and third (90^{th} percentile) highest copper concentration for each alkalinity tested. Additional models are presented including Mineql+ based on equilibrium with $Cu_3(PO_4)_2$ solid and empirical predictions based on Lytle et al.,[206] with varying pH corresponding to those used to develop "minimum" orthophosphate criteria.

B.2 Mineql+ Modeling of cuprosolvency

Cuprosolvency modeling was carried out using Mineql+ software (version 4.6) and assuming the system was in equilibrium with $Cu_3(PO_4)_2$ solid (13 °C). First, we calibrated the model to our data by adjusting the logK to fit our data at pH 7.5 and 100 mg/L as $CaCO_3$ alkalinity. The rest of the Mineql+ solubility models were run using the adjusted logK (37.6). Titration mode was used for each pH and alkalinity to determine the soluble orthophosphate residual concentration that would be needed to yield a soluble copper concentration of 1.3 mg/L.

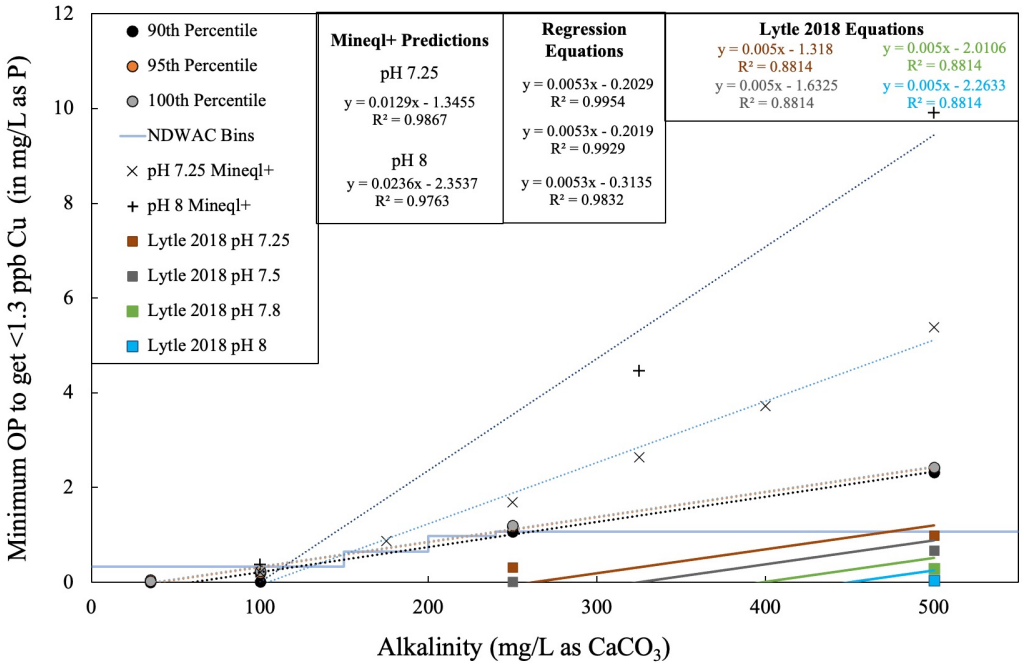


Figure B.3: **Linear regressions defining the "minimum" orthophosphate criteria** for varying alkalinities to define waters "non-aggressive" to copper. Orthophosphate values represent the highest interpolated value, after either 4 or 22 weeks of testing, from linear correlations of orthophosphate and maximum (100^{th} percentile), second (95^{th} percentile) and third (90^{th} percentile) highest copper concentration for each alkalinity tested. Additional models are presented for comparison including Mineql+ predictions at pH 7.25 and 8 for cuprosolvency in equilibrium with $Cu_3(PO_4)_2$ solid as well as empirical predictions based on Lytle et al.,[206] at pH 7.25, 7.5, 7.8, and 8.

B.3 Estimation of cuprosolvency testing costs

Three cuprosolvency tests were compared based on cost and time investment. All estimates assume access to ICP/MS-MS capabilities ($6 per sample), access to a working pH meter, labor costs of $20/hr., and use of utility or residential test waters. Estimates for cuprosolvency tests using copper tube and fresh copper solids reflect approximate labor and expenses from test in our laboratory. All estimates are for materials from Fisher Scientific or Home Depot, unless otherwise noted. Many of these costs would be similar for other laboratories or utilities trying to carry out similar experiments, however, some costs may vary based on sample analysis costs and where materials are sourced from.

Estimates for tests using fresh copper solids include initial one-time costs such as buying a carboy (10L, $137), plastic bottles (125 mL, two 12 packs, $93), and a stir plate with rpm control (10.25", $849), all of which could be reused in subsequent trials. However, many consumables would be needed including: reagents (trace metals grade nitric acid, 500 mL, $180), syringes (10 mL, 50 pack, $56), syringe filters (150 pack, 0.45 µm, $839), disc filters (100 pack, 0.45 µm, $345), and sample analysis ($6 per sample for ICP-MS/MS analysis). The experiment was estimated to take 12 non-consecutive hours for initiating the experiment and collecting and analyzing samples.

Estimates for cuprosolvency tests in copper tube include one-time costs for a carboy (10L, $137), plastic bottles (125 mL, two 72 packs, $470), a pipe reamer ($15), materials for a storage tray ($16) and weighted lid ($58, including foam mat, metal sheet, glue, and metal pieces for weight), and a wine refrigerator for temperature control ($729), all of which could be reused in subsequent trials. In addition, these tests require consumables including copper tube (2 10' pieces cut into 8.5" segments, $50), silicone stoppers (Cole Parmer 13D, 2 10 packs, $42), reagents (trace metals grade nitric acid, 500 mL, $180), sample analysis ($6 per

sample for ICP-MS/MS analysis), and additional supplies ($26, including cups, plastic wrap, foil, rubber bands, labels). Finally, the experiment was estimated to take 80 non-consecutive hours, including procuring materials, preparing pipes (cutting, reaming, and cleaning), water changes three times per week, weekly composite sampling, monthly sampling of individual pipes, and sample preparation and analysis.

Estimates for in-home testing assumes a visits by staff to a resident's home to perform sampling using a 3-bottle sequential sampling approach. Costs could vary widely depending on whether residents could sample themselves, how often sampling took place, and for what duration. We assumed sampling one time with the sampling taking total of 6 hours of utility personnel time. This labor includes time related to transit and sample collection in the home as well as sample preparation and analysis. In addition, this sampling would require 3 1L bottles ($84) as well as reagents (trace metals grade nitric acid, 500 mL, $180) and sample analysis ($6 per sample for ICP-MS/MS analysis).

Estimates also determined the approximate cost of running additional tests. These estimates assumed the same labor and analysis costs as previous tests. To account for additional materials that may be left over from previous tests, these estimates do not include costs for syringes, syringe filters, disc filters, nitric acid, or cupric nitrate reagents. Further, these estimates include an additional 2 hours of labor for In-Home and Fresh Solids Tests and an additional 5 hours of labor for Copper Tube Tests to account for disposing of samples and cleaning bottles for reuse.

Appendix C

Supplemental Information for Chapter 4

C.1 Determination of "minimum" pH criteria

The "minimum" pH values needed to achieve copper concentrations below the 1.3 mg/L action level were estimated based on water alkalinity. To determine this, three pH values were tested for each of four alkalinities (i.e. 12 water conditions, each with n=20 pipe segments). Results for the 90^{th}, 95^{th}, and 100^{th} percentile (third highest, second highest, and highest) copper values were considered after 4, 22, and 35 weeks of tube/pipe conditioning (11 weeks with water changes and extended stagnation periods of 14-weeks and 10-weeks due to COVID-19 lab shutdowns). Results for the 90^{th}, 95^{th}, and 100^{th} percentile copper were determined to facilitate policy considerations such as use of a 90^{th} percentile AL under the current LCR.

This section provides an example of the method used to determine the "minimum" pH criteria by considering the 90^{th} percentile copper concentrations (presented in Figure C.1). The 90^{th} percentile copper concentrations represent the third highest copper concentration of n=20 pipes. This section will focus on determining "minimum" criteria at 35 mg/L as $CaCO_3$ alkalinity.

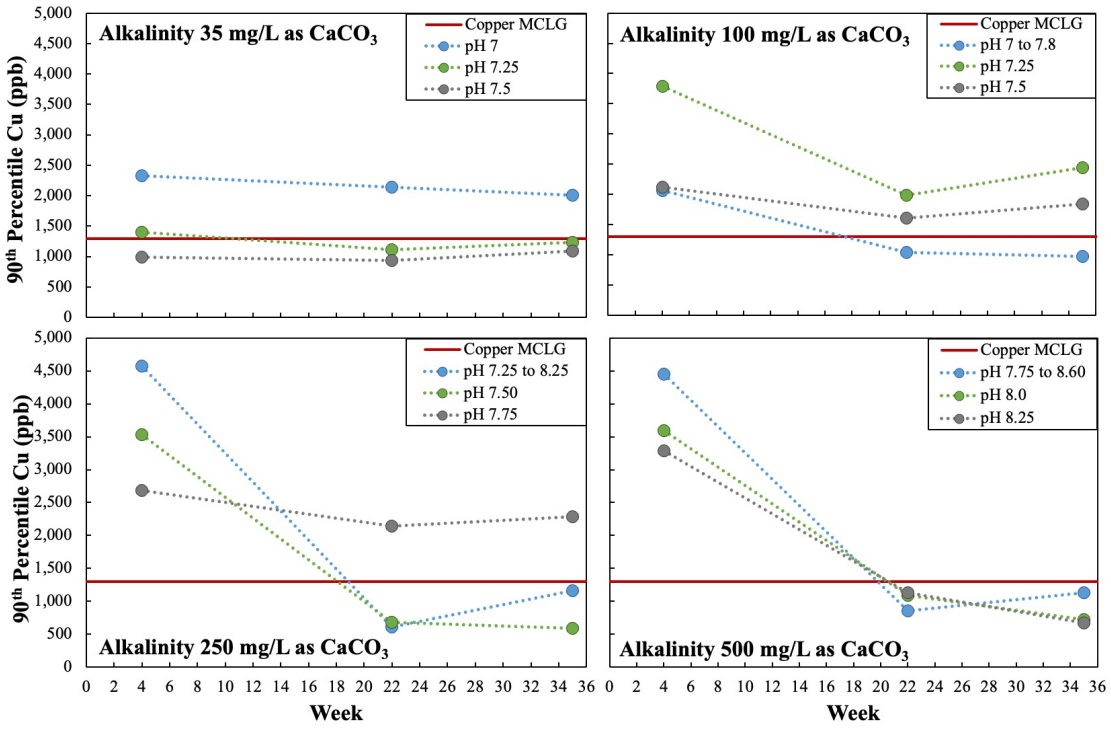


Figure C.1: **90th percentile copper release from new copper tube treated with varying pH** (n=20 for 4, 22, and 35 weeks). Red line represents the 1,300 ppb US EPA AL.

First, we consider only 90th percentile results at 35 mg/L as $CaCO_3$ alkalinity after 22 weeks of aging. Interpolating the pH vs. resulting 90th percentile copper concentration can be used to estimate the pH needed at 35 mg/L as $CaCO_3$ alkalinity and 22 weeks of aging that would yield copper at or below the 1.3 mg/L AL (Figure C.2). Under these conditions a pH of 7.23 is predicted to yield copper below the AL.

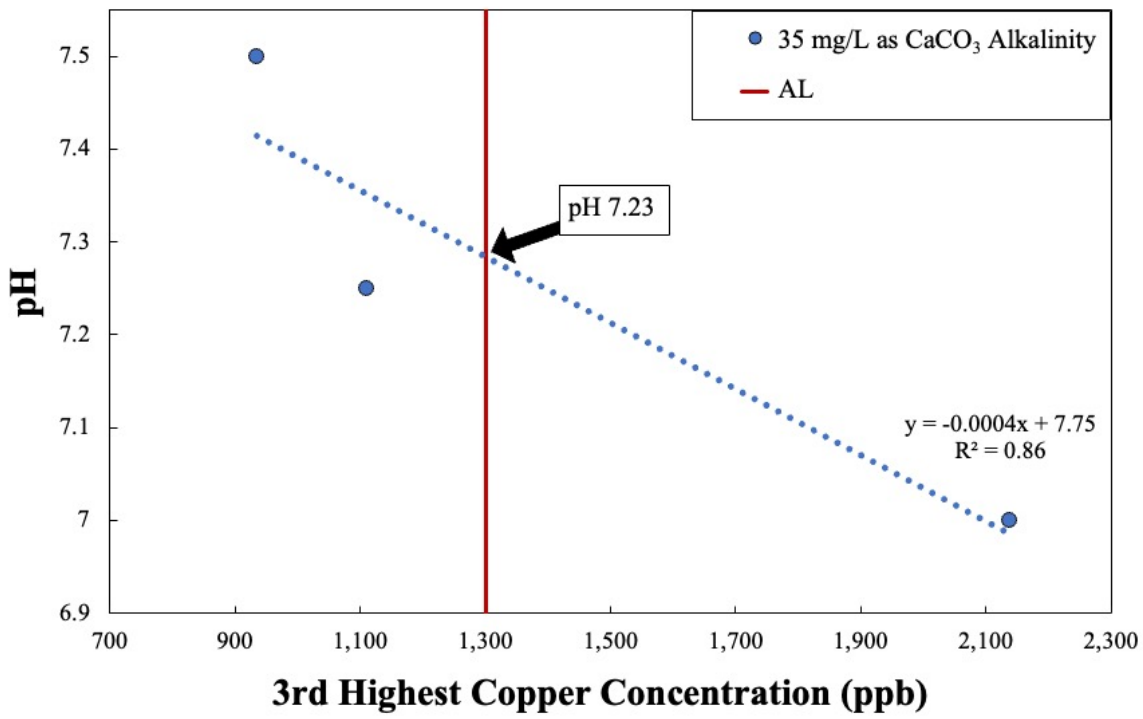


Figure C.2: **Illustrative interpolation for determining the minimum pH value needed to achieve copper below the Action Level for the 90th percentile (third highest) copper concentration after 22 weeks of pipe aging.** The arrow with box denotes the "minimum" pH value predicted by the regressions to yield copper below the action level.

This process can be repeated for the 35 mg/L as $CaCO_3$ water conditions at the other two time periods (4 and 35 weeks of pipe aging). These interpolated results indicate that a pH of 7.29 would be needed after 4 weeks of aging, a pH of 7.23 would be needed after 22 weeks of aging, and a pH of 7.28 would be needed after 35 weeks of aging to yield copper below the AL. In order to provide a conservative estimate for the "minimum" pH criteria, the highest of these three values (i.e. pH 7.29) was used as the "minimum" pH for the 90th percentile copper at 35 mg/L as $CaCO_3$ alkalinity in development of the linear regression presented in Figure 4.1A. The same method was repeated to determine the "minimum" pH criteria for the 90th percentile copper concentrations at the other three alkalinities and for the 95th and

100^{th} percentile copper concentrations at all four alkalinities.

C.2 Solid phase identification from fresh-solids cuprosolvency tests

Solid phases were identified via XRD by a collaborator at the US Environmental Protection agency. Results indicate that most solids formed by the end of fresh-solids cuprosolvency tests were crystalline malachite, with a few conditions having tenorite or cupric sulfates (brochantite, posnjakite). Several high alkalinity conditions with added sulfate did not form cupric sulfates, indicating that other solids like tenorite and malachite were favored at high alkalinity conditions. Unlike other samples yielding uniform crystalline solids, many silica conditions yielded poorly crystalline solids largely made up of Cu, O, and Si, that flaked off filters when dried. The combination of poorly crystalline solids and observed higher long-term cuprosolvency indicate that silica may affect formation of protective scales in new copper pipes, as observed by Nguyen et al.[231]

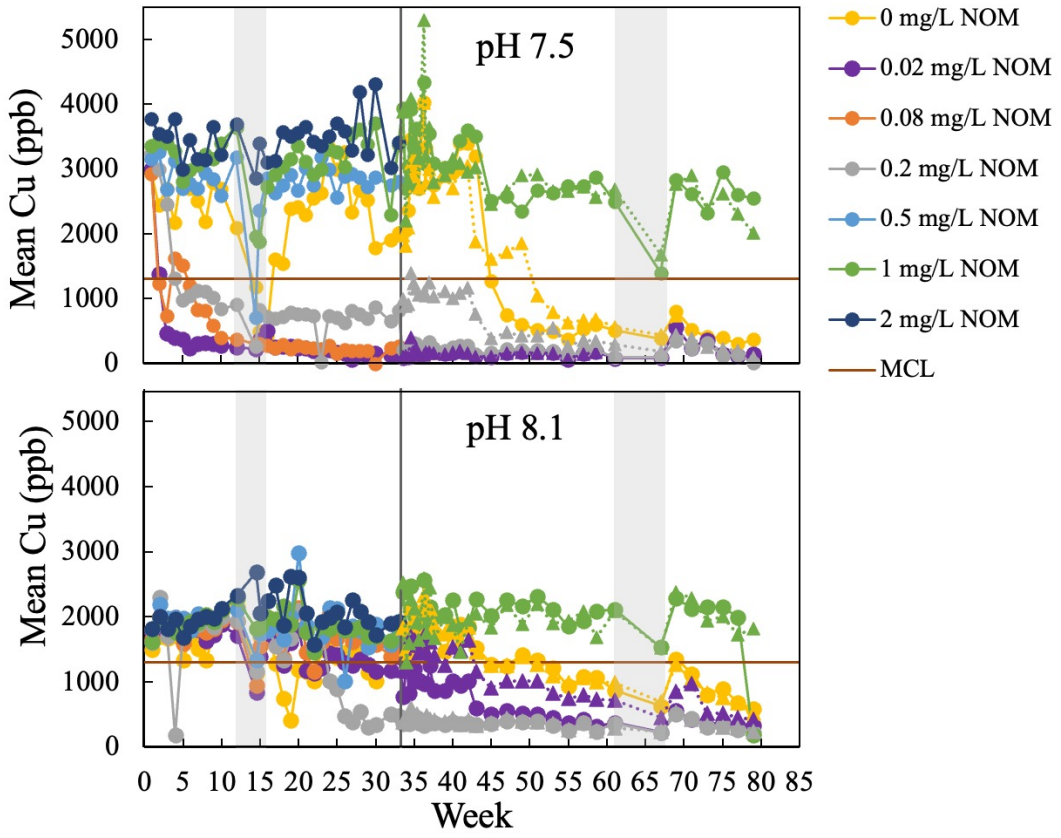


Figure C.3: **Effects of NOM on cuprosolvency for pH above and below predicted pH criteria.** Copper tube segments (n=5, one manufacturer). Vertical line (week 33) represents switch in water conditions from 200 mg/L sulfate (circles) to 0 mg/L sulfate (triangles) for the 3 pipes with the highest copper in each water condition. Grey bars represent longer stagnation periods than normal.

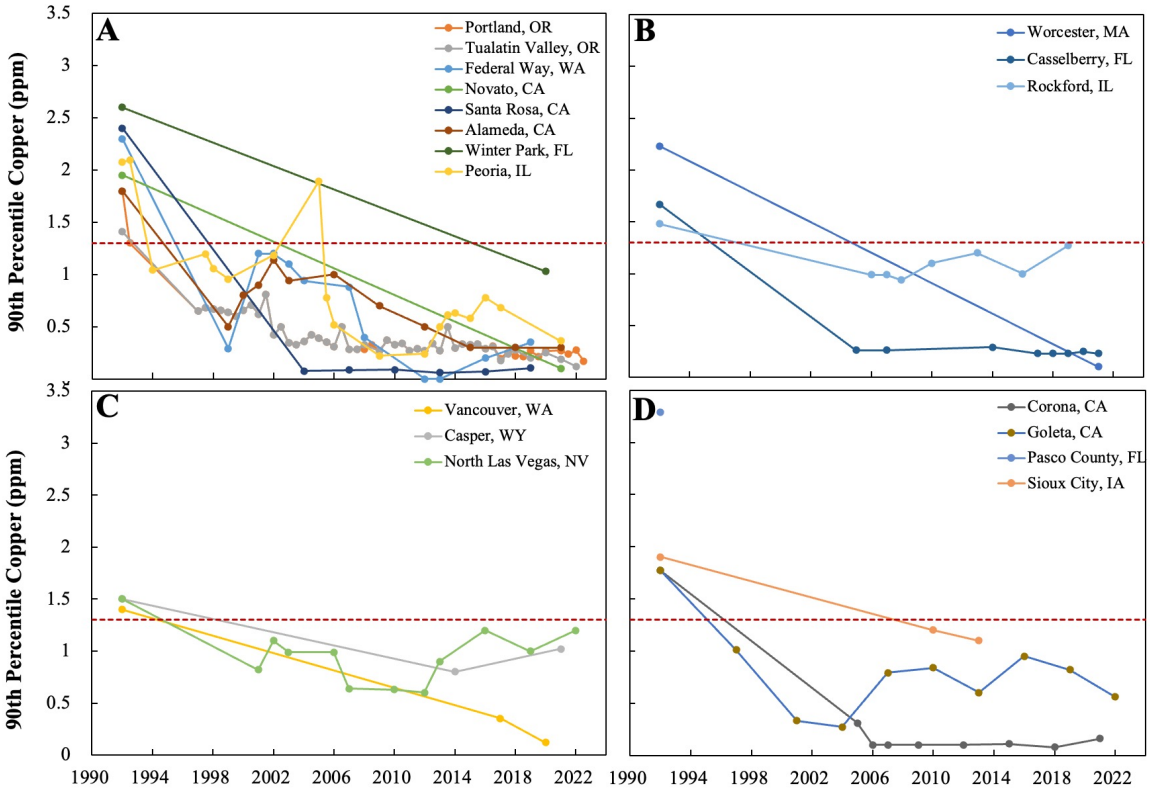


Figure C.4: **Utility LCR monitoring data for copper based on CCT.** Utility 90$^{\text{th}}$ percentile copper concentrations collected in LCR monitoring are presented from 1992 to present based on corrosion control: A. pH-based CCT, B. Phosphate-based CCT, C. Unspecified CCT, and D. No CCT

Appendix D

Supplemental Information for Chapter 5

Table D.1: **Comparison of in-home sampling and pipe segment cuprosolvency results from the home of Resident A**

			Home Plumbing		Pipe Segments	
Location	Time	Sample Type	Cu (mg/L)	Current (Microamps)	Avg. Cu (mg/L)	SD
Kitchen	9:00 PM	Flushed (5 min)	0.2			
	7:00 AM	1st Draw	0.5	103	1.1	0.1
	7:00 AM	20 sec. flush	0.9	103		
Hose Bib	9:00 PM	Flushed (1 min)	0.1			
	7:00 AM	1st Draw	0.9	98	1.1	0.0
	7:00 AM	20 sec. flush	1.0	98		
Nextdoor	9:00 PM	Flushed (1 min)	0.1			
	7:00 AM	1st Draw	0.0	135		
	7:00 AM	20 sec. flush	0.1	135		

Appendix E

Understanding lead in water and avoidance strategies: a United States perspective for informed decision-making

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https://iwaponline.com/jwh/article/17/4/540/67697/Understanding-lead-in-water-and-avoidance

Abstract

The pervasiveness of lead in drinking water poses a significant public health threat, which can be reduced by implementing preventive measures. However, the causes of elevated lead in water and the benefits of lead in water avoidance strategies are often misunderstood. Based on experiences in the United States, this paper describes an oversimplified 'lead in water equation' to explain key variables controlling the presence of lead in drinking water to better inform public health practitioners, government officials, utility personnel, and concerned residents. We illustrate the application of the equation in Flint, Michigan and explore the primary household-level water lead avoidance strategies recommended during the crisis, including flushing, filtration, bottled water use, and lead pipe removal. In addition to lead reduction, strategies are evaluated based on costs and limitations. While these lead avoidance strategies will reduce water lead to some degree, the costs, limitations, and effectiveness of these strategies will be site- and event-specific. This paper presents a simplified approach to communicate key factors which must be considered to effectively reduce waterborne lead exposures for a wide range of decision makers.

Key words | corrosion, Flint Water Crisis, lead avoidance strategies, lead in drinking water

E.1 INTRODUCTION

The short- and long-term impacts of chronic exposure to low doses of lead from drinking water are still being investigated, but the U.S. Centers for Disease Control and Prevention (CDC) has asserted that no amount of exposure is safe. Given the bioaccumulative nature of lead, the lack of a health-based exposure threshold, and the U.S. Environmental Protection Agency's (USEPA) acknowledgement that drinking water regulations may not protect all individuals from potentially harmful lead exposures, exposure prevention is paramount[226]. Drinking water is considered, by some, to be an underestimated source of lead, due to the corrosion of leaded drinking water infrastructure, lack of adequate treatment methods for controlling particulate lead, and sample collection methods that can undermine monitoring purposes[107, 298, 334]. Thus, instead of reacting to non-health-based regulatory triggers, public health officials should encourage vulnerable populations to take proactive precautionary measures to prevent water lead exposure and empower people with information on evidence-based strategies and technologies. Such evidence-based interventions are needed for both acute exposures such as those observed during the Flint Water Crisis and low-dose chronic exposures that are common in many Lead and Copper Rule (LCR) compliant cities with lead service lines (LSLs)[172, 259].

While the Flint Water Crisis has elevated national concerns about water lead exposure[142], there have been other high profile contamination events that caused serious childhood lead exposure.[109, 112, 214] For instance, the 2001–2004 Washington DC Lead Crisis went largely unreported to the public for 4 years, caused hundreds of cases of elevated blood lead over the old CDC level of concern (10 µg/dL), and was associated with increased fetal death and miscarriage rates[107, 117]. Although a perfect comparison is not possible due to differences in sampling pools and methods, available data (Figure E.1) suggest that the Washington DC

first draw water lead levels (WLLs) were much higher than Flint ([117, 259]. However, it is important to note that first draw samples (i.e., samples collected after 6+ h of stagnation) do not always represent the worst-case WLLs, as these samples do not adequately characterize lead release in the particulate form or from LSLs[70, 84, 109, 172, 255, 258]. Learning from these and other water lead contamination events can help avoid future problems during water crises, action level exceedances, and harmful exposures that occur in cities that meet the LCR.

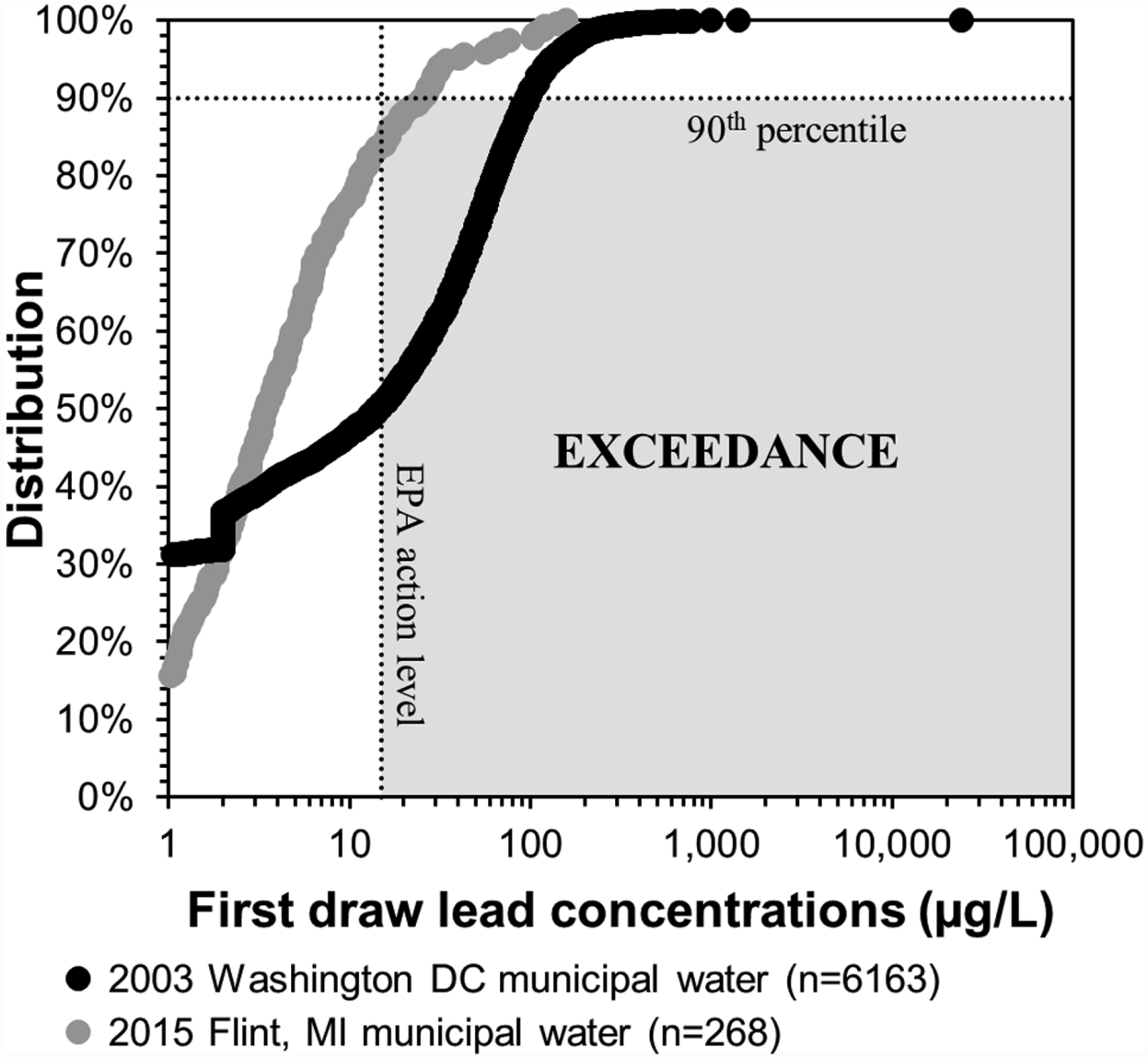


Figure E.1: **Representative histogram of WLLs during the Flint and Washington DC lead in water crises**[117, 259].

The US EPA enacted the 1991 LCR to prevent wide-spread water lead exposure by reducing water corrosivity through corrosion control treatment (CCT) — corrosion control reduces the propensity of water to pick up lead when contacting plumbing infrastructure[344]. According to the LCR, water utilities must conduct the limited monitoring of high-risk homes under normal residential use conditions. High-risk homes are determined based on the age of

structure and plumbing material composition, rather than selecting sites based on zonal variations in water corrosivity. If more than 10% of first draw samples exceed the lead action level of 15 µg/L, water utilities must optimize CCT, collect additional water samples, and notify the public. However, satisfying the LCR's action level requirements does not guarantee that a city's tap water is free of lead and is safe for all residents to consume[171, 344]. The Natural Resources Defense Council revealed that in 2015, 5,363 water systems, serving more than 18 million US residents, had LCR health, monitoring, and/or reporting violations[239]. USA Today documented high WLLs in 350 schools and day-care centers between 2012 and 2015[341], and also reported that 9,000 small water systems, serving almost 4 million rural residents, failed to test for lead in the past 6 years[342]. Moreover, private well users are not protected under the LCR, as private water systems (e.g., wells, springs, and cisterns) are not regulated by the USEPA[344].

E.2 CONCEPTUAL LEAD IN WATER EQUATION

The three key variables that influence the presence of lead in drinking water at homes are: (1) lead-bearing plumbing; (2) corrosive water; and (3) ineffective CCT (Figure E.2). The worst-case combination of these variables will produce the highest levels of lead in drinking water, whereas correcting one or all of these variables can potentially reduce or prevent lead in drinking water. The equation is qualitative rather than quantitative and underscores factors that must be considered when addressing lead in water issues.



Figure E.2: **Oversimplified conceptual equation illustrating variables leading to lead in drinking water.**

E.2.1 Lead-bearing plumbing

The use of lead in plumbing materials has been reduced over the years through USEPA regulations and industry best practices (Figure E.3; Table F.1). The 1986 Safe Drinking Water Act Amendments banned leaded and pure lead plumbing by requiring the installation of 'lead-free' plumbing[4]. However, 'lead-free' plumbing materials could still contain lead – up to 8% by weight until 2014 and a weighted average of 0.25% based on wetted surfaces thereafter[37].

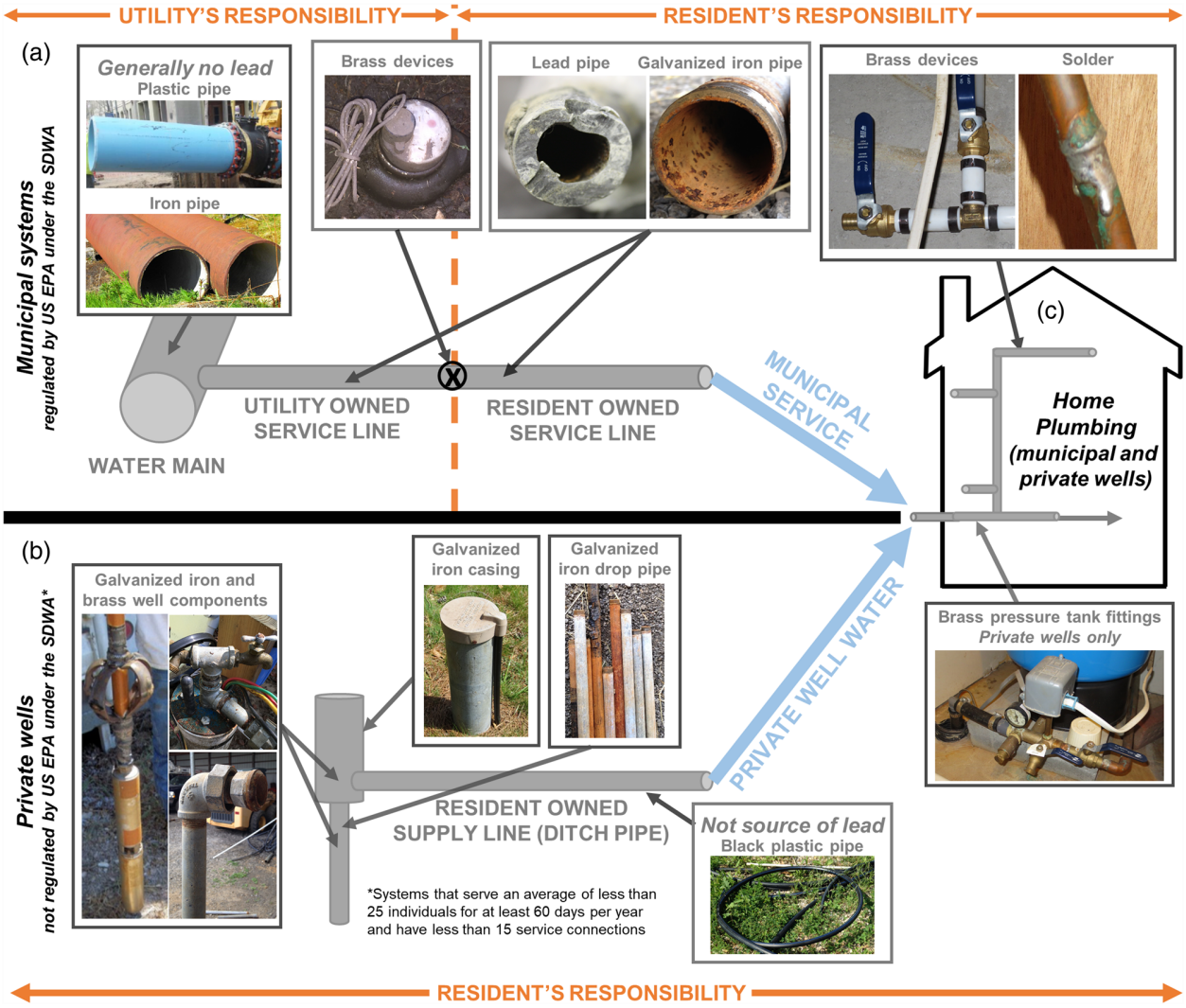


Figure E.3: **Lead-bearing plumbing components potentially used within drinking water systems.** (a) Municipal water systems with responsibility split between the utility and the resident; (b) private wells which are solely the responsibility of the resident; and (c) home plumbing system which are solely the responsibility of the resident.

E.2.2 Lead-bearing service line materials

Pure lead pipes were widely used for service lines (pipe connecting premise plumbing and water main; Figure E.3(a)) until the 1950s, though some cities continued to use LSLs until these pipes were banned in 1986[271]. LSLs are, by far, the most concentrated source of lead

present in homes (100% lead) and can directly contaminate drinking water. When present, LSLs are often responsible for 50–75% of lead observed at the tap[291]. Replacing LSLs as a solution to mitigate lead sources can sometimes be problematic as ownership of this pipe is split between the homeowner and water utility. When homeowners do not replace their section of the service line, only part of the lead pipe is replaced, which results in a partial lead service line replacement (PLSLR). When the utility's portion of the lead pipe is replaced with the copper pipe or brass connector fittings are installed, higher levels of lead sometimes result from accelerated corrosion of the lead pipe[64, 291, 316, 334] and physical disturbances of leaded scales may occur[84]. As a result, occasionally PLSLRs do not reduce short- or long-term WLLs, but can even cause worse water lead issues.

Although LSLs are the primary service line material of concern, galvanized iron service lines (Figure E.3(a) and well components (Figure E.3(b)) can also serve as a lead source and have been attributed to water lead problems[71, 258, 259, 260, 291]. A galvanized iron pipe is an iron pipe with a protective zinc–lead 'galvanized' surface coating. This galvanized coating often contained between 0.5 and 1.4% lead by weight until 2014[71]. Lead can leach into water from a pre-2014 galvanized iron pipe and be distributed to the tap or accumulate in iron rust layers along the interior of the pipe – creating both short- and long-term water lead problems[71, 258, 260]. Even new post-2014 'lead-free' galvanized iron pipes (<0.25% lead in the surface coating) are still of concern due to the potential formation and remobilization of leaded rust scales[257].

E.2.3 Lead-bearing household plumbing materials

Lead-bearing plumbing materials are still commonly used in home construction (Figure E.3(c)). Leaded solders (composed of 40–50% lead) were used to connect copper plumb-

ing until 1986[4, 112, 334]. Several cases of childhood lead poisoning have been directly linked to the detachment of pieces of leaded solder[112, 335]. More recently, there have been concerns regarding pre-2014 'lead-free' brass (<8% lead by weight), as this lead-bearing component has been linked to several incidences of high WLLs in schools, buildings, and homes[120, 176, 205, 256, 334]. Regardless of the piping material (copper or plastic), brass fittings such as ball valves, elbows, and faucet components are often present in the premise plumbing. Fortunately, 'lead-free' components containing <0.25% lead by the wetted surface[37] have become available, which release lower lead in water[257] However, new recommendations by the American Association of Pediatrics to reduce water lead to below 1 µg/L in school water may be difficult to achieve even with some products designed to meet the 2014 'lead-free' standard[77, 249].

E.2.4 Water Corrosivity

Some waters are naturally corrosive, whereas other waters are naturally non-corrosive. There are several well-established water chemistry parameters that influence the corrosivity of drinking water such as dissolved oxygen, pH, water disinfectants, chloride-to-sulfate mass ratio, and alkalinity[297, 298, 333]. These parameters are controlled and routinely monitored by the drinking water operators (see E.2.4). Although lead cannot typically be detected by taste, smell, or sight in water, studies note that higher water corrosivity (and resulting WLLs) can sometimes associate with certain unpleasant or undesirable characteristics of the drinking water. For example, private well users who had obvious signs of corrosion (e.g., plumbing leaks), blue-green staining on plumbing fixtures, and described the taste of water as metallic were more likely to have copper concentrations and low water pH, which were correlated with high WLLs[256]. Researchers have also occasionally linked the incidence of red/rusty water reports to elevated WLLs, as the corrosion of iron pipes may indirectly result

in higher WLLs[214, 258, 259]. While the presence of red/rusty water may be an indicator of lead in some situations, that is often not always the case[327].

E.2.5 Ineffective CCT

Corrosion control by public water supplies involves the manipulation of pH, the adjustment of alkalinity, and/or the addition of corrosion inhibitors (e.g., phosphates or silicates), to reduce problems from lead pipes and other plumbing. However, residents reliant on private wells are responsible for implementing corrosion control, with associated responsibilities for monitoring and maintenance[256, 324]. Although corrosion chemistry can be complex and dependent on plumbing materials (e.g., brass and solder), appropriate CCT can reduce lead in water through three dimensions of performance: (1) minimizing the dissolution of soluble lead and leaded scale layers by adding corrosion inhibitors to the water or increasing water pH and/or alkalinity (Figure F.1(a)); (2) promoting the development of protective scale layers that reduce corrosion rates and the dissolution of soluble lead (Figure F.1(b)); and (3) increasing the durability of leaded scale layers to prevent the destabilization and detachment of such layers to water (Figure F.1(c))[110, 297]. According to the LCR, any water utility serving 50,000 residents must have a state-approved optimized CCT plan[344]. CCT is only required in systems serving <50,000 residents when a utility exceeds the lead and/or copper action level during their required water sampling. The installation of CCT devices in private wells is limited and only corrects the water chemistry after treatment and only rarely the chemistry of water within the well plumbing[255, 256, 324].

E.2.6 Measuring lead in water

The LCR requires the collection of first draw samples, which was once considered the worst-case scenario as dissolved lead concentrations increase with stagnation time. This 1 L first draw sample will typically capture 7.9 m (25.9 ft) of a 6.4 mm (0.25 in.) diameter pipe, which only includes house-hold (or premise) plumbing and not lead from the service line. Researchers have concluded that single first draw samples may not capture the worst-case water lead, which is particularly true for homes with LSLs or homes with particulate lead problems[70, 84, 109, 172, 255, 258, 273]. Other factors that can result in reduced detection of lead hazards include sampling in cold weather and at low flow rates, prior removal of aerator filters, pre-flushing the night before collection, and inadequate sample processing in the laboratory[171]. In recognition of inherent weaknesses in the LCR sampling protocol, researchers are utilizing profile sampling methods to more accurately characterize the entire plumbing network, including the premise plumbing and service lines, and its detachment in response to higher flow rates[70, 84, 255, 258]. Thus, it is imperative that proper sampling protocols be used to quantify water lead accurately.

E.2.7 Study Objectives

The pervasiveness of lead in drinking water poses a significant public health threat, but exposure can be reduced or prevented almost completely, through the implementation of preventive measures. However, problems with monitoring, regulating, and remediating water lead have long been misunderstood due to the complexity of plumbing and corrosion control. With the USEPA's new 'war on lead'[312], it is imperative that simple but accurate scientific information be communicated effectively to a wide range of decision makers to reduce water lead exposures. This paper illustrates the application of an oversimplified 'lead

in the water equation' to explain the key variables that control the presence of lead in water to lay audiences and presents factors to consider when selecting a household-level water lead remediation strategy. A case study methodology is used to inform practice based on the Flint Water Crisis. This work aims to provide public health practitioners, government officials, utility personnel, and concerned residents with a science-based model to inform communications, decision-making, and implementation of household-level avoidance strategies for lead from drinking water.

E.3 METHODS

The application of the lead in the water equation is demonstrated through a case study of Flint, Michigan. Published and newly collected data from Flint were applied to evaluate the four primary water lead avoidance strategies: (1) flushing, (2) bottled water, (3) lead filters, and (4) LSL replacement (Table E.4 E.5). The overall approach was to evaluate each strategy based on costs, limitations, and reductions in lead exposure (defined as WLL reductions or by water lead avoidance).

Figure E.4: **Comparison of the four household-level lead remediation strategies used during the Flint Water Crisis.** [a] Intervention costs (bottled water, filters, and LSL replacements) were provided to Flint residents at no cost at times during recovery. [b] Cost of water in Flint[272]. [c] Water lead measured in April 2015[258]. [d] Water lead measured in August 2015 and March 2016[259]. [e] Measurements and data collected during this effort. [f] Filter assessment at Virginia Tech in February 2016[208]. [g] Filter assessment by USEPA in January 2016[19].

		Reduction in WLLs (µg/L)			Financial costs[a]		
		Initial water lead value	Intervention water lead value	Percent of samples reduced <AL	Cost of water per gallon[b]	Installation and maintenance fees	Non-exhaustive tangible and intangible impacts Potential impacts in Flint
Flushing	*Home of resident zero[c] (1 home; 32 samples)*	First draw of 2,171 µg/L	3,550 µg/L *at 1 min* 1,412 µg/L *at 3 min* 2,542 µg/L *at 5 min* 1,742 µg/L *at 25 min*	0%	<$0.001	*Volume of water flushed* 1 min: 8.3 L (2.2 gal) 3 min: 24.9 L (6.6 gal) 5 min: 41.5 L (11.0 gal) 25 min: 207.5 L (55.0 gal) *Price of water flushed* $0.002 per 1-min flush $0.006 per 3-min flush $0.01 per 5-min flush $0.05 per 25-min flush	*Extra water use burden on water utility with morning and afternoon 3 min flush* Daily: 2.2 million L (0.6 million gal) 30 days: 65 million L (17 million gal) *Extra water use burden on household with morning and afternoon 3 min flush* Daily: $0.01 30 days: $0.35 ***Other considerations:*** - Cost of water for local utility - Water scarcity challenges - Water and wastewater treatment burden - Infrastructure impacts - Additional demand - Educating residents on the protocol - Prompting intervention adoption - Developing education materials - Perception of water safety
	Community-wide in August 2015[d] (268 homes; 3 samples per home)	90th percentile of 26.8 µg/L	11.3 µg/L *at 1 min* 6.6 µg/L *at 3 min*	94% 96%			
	Community-wide in March 2016[d] (156 homes; 3 samples per home)	90th percentile of 22.4 µg/L	9.0 µg/L *at 1 min* 3.2 µg/L *at 3 min*	96% 99%			
Bottled water (costs and water lead estimates derived from three samples from each of five brands)[e]		–	<1 µg/L	100%	$0.77–$8.32[e]	*Family of four drinking and cooking water needs* Daily: 5 L (1.3 gal) 30 day: 600 L (159 gal) *Price of bottled water* 30 day: $122.05 Save $0.14 on water bill	*Plastic bottle to solid waste* Daily: 5–40 bottles 30 days: 159–1,200 bottles ***Other considerations:*** - Ongoing cost of bottled water - Reduced water use from utility - Resident may use less water (7 gal/day compared to 100 gal/day) - Transportation to procurement/ distribution of bottled water - Trash/recycle burden - Environmental and utility burden of discarded materials - Benefit of the avoidance of other contaminants and taste/odor compounds - Educating residents on bottled water use - Prompting intervention adoption - Developing education materials

Figure E.5: **Comparison of the four household-level lead remediation strategies used during the Flint Water Crisis. (Continued)**

[a] Intervention costs (bottled water, filters, and LSL replacements) were provided to Flint residents at no cost at times during recovery.

[b] Cost of water in Flint[272].

[c] Water lead measured in April 2015[258].

[d] Water lead measured in August 2015 and March 2016[259].

[e] Measurements and data collected during this effort.

[f] Filter assessment at Virginia Tech in February 2016[208].

[g] Filter assessment by USEPA in January 2016[19].

		Reduction in WLLs (µg/L)			Financial costs[a]		
		Initial water lead value	Intervention water lead value	Percent of samples reduced <AL	Cost of water per gallon[b]	Installation and maintenance fees	Non-exhaustive tangible and intangible impacts Potential impacts in Flint
NSF 53 filters	*Home of Resident Zero[f] (1 unfiltered and 1 filtered sample)*	Influent of 13,200 µg/L	20 µg/L	0%	< $0.001	*Installation*: $15–50 for filter unit *Maintenance:* $10–15 per cartridge	***Other considerations:*** - Ongoing cost of filter replacements - Burden of getting initial and replacement filters - Environmental and utility burden of discarded filters - Benefit of the avoidance of other contaminants and taste/odor compounds - Educating residents on the protocol - Prompting intervention adoption - Developing education materials - Perception of water safety - Perceived risk of microbial contamination
	Community-wide[g] (241 homes; 1 unfiltered and 1 filtered sample per home)	Unfiltered 90th percentile of 68 µg/L	Filtered 90th percentile of <1 µg/L	100%		Tap-mounted: replace filters every 3–4 months Pitcher style: replace filters every 1–2 months	
LSL replacement[e] *(1 house; 18 samples)*		First draw of 2,171 µg/L[c]	2.1 µg/L *first draw* <1 µg/L *at 1 min* 32.4 µg/L *at 2 min*	94%	<$0.001	Estimated $2,800 to replace 25 ft. LSL and 192 ft. galvanized iron service line	***Other considerations:*** - Perception of safety - One-time high cost to utility - Cost to consumer for full LSLR - Removal, transportation, and disposal old materials

E.3.1 Flushing

Data from two publications[258, 259] were used to evaluate reductions in WLLs. Based on an average kitchen faucet water flow rate of 8.3 L/min (2.2 gal/min)[361], the volume of water flushed was calculated at the household and city level. The price of water in Flint used in this assessment was from a 2016 Raftelis Financial Consultants (RFC) report, which was $3.30 per 14,195 L (3,740 gal)[272]. This rate was used to evaluate the increase in water bills at the household and city level.

E.3.2 Bottled Water

To determine WLLs in bottled water, five brands distributed during the Flint Water Crisis (Deer Park, Great Value, Kroger, Member's Mark, and Nestle) were analyzed. Specifically, a 10 mL aliquot was collected from each bottle after thorough shaking, and three bottles of each brand were analyzed (n = 15 samples; three bottles from five brands). To evaluate bottled water costs, we reviewed popular brands available in three Virginia grocery stores in August 2018. Using reported statistics on average water use per person, it was assumed that 5 L (1.3 gal) was the volume of water used per day for all cooking and drinking purposes[21]. Lastly, the savings from switching from municipal water to bottled water was calculated based on water volumes and the RFC report.

E.3.3 Lead Filters

The USEPA published data from their in-home filter efficacy testing[19]. For our analysis, only homes and buildings with paired unfiltered samples and filtered samples collected after the installation of a new filter were used (n = 241). It is important to note that unfiltered

samples were not required to have 6+ h of stagnation until March 2016, and filtered samples were collected after running water through the new filter for 2 min. If multiple unfiltered samples were collected, samples were collected with the aerator removed (n = 5) and first draw samples (n = 2) were selected. Paired samples collected from the same home (n = 20) were used in this analysis if the samples were collected from different faucets and/or on different sampling dates with a new filter being installed. The minimum reporting limit for lead during the USEPA study was 0.5 µg/L. To evaluate filter costs, we reviewed popular brands available in four national box stores in August 2018.

E.3.4 LSL Replacement

To evaluate WLL reduction after replacing the service line at the home of a Flint resident with high sustained WLLs[258], follow-up profile samples were collected. Specifically, after 6+ h of stagnation, 18 sequential 1 L samples were collected at normal flow and shipped to Virginia Tech for analysis. All samples and aliquots were acidified with 2% (v/v) concentrated nitric acid and digested for a minimum of 16 h before analysis using inductively coupled plasma mass spectrometry (ICP-MS) per method 3125 B[72]. For data quality assurance and quality control, blanks and spikes of known concentrations were measured every 10–15 samples. The costs of a full or partial line replacement depend strongly on site-specific considerations, such as access constraints, but cost estimates have ranged from $1,000 to $7,000 per home[187] to $2,500 to $8,700 per home[20].

Limitations of cost and cater lead estimations

There are limitations to the approaches used to estimate costs and WLL exposures. Indirect costs (e.g., increased solid waste and recycling of plastic bottles) were not included in this

analysis, but they are important to identify and quantify[358]. Also, WLLs can be highly variable, which makes realistic measurements of exposures to lead in water difficult to calculate. The focus of this paper was a case study for Flint, Michigan, and it is advisable that when this framework is applied to other locations, decisions should be based on as much site-specific data as are available. Thus, this framework aims to help communities consider and evaluate appropriate intervention strategies.

E.4 RESULTS

E.4.1 Using the lead in the water equation in Flint

This section presents how the lead in the water equation (Figure E.2) can be applied through a case study of the lead water crisis in Flint, Michigan. In Flint, all three factors were involved, resulting in high lead in water levels.

Lead-bearing Plumbing

Most housings in Flint were constructed between the 1950s and 1960s, and few homes were built after 1986, suggesting a high prevalence of LSLs and lead solder. Previously, the Michigan Department of Environmental Quality (MDEQ) estimated that at least 15,000 service connections were full or partial LSLs[14]. New estimates suggest that 29,100 service line connections are either lead or galvanized iron service lines[220].

Corrosive Water

The Flint River water had different water chemistry (e.g., chloride levels and pH), resulting in a drinking water supply that was more corrosive to the drinking water infrastructure than Detroit water[83, 95, 258]. Specifically, when exposed to Flint River water, metal release into drinking water was 3.5–4.2 times higher[95]. This increase in water corrosivity was also evident in the deterioration of water quality, as residents reported changes in taste, smell, and clarity of the water (Figure F.2)[60, 123].

Ineffective CCT

When switching to treated Flint River water, city officials did not continue adding orthophosphate inhibitors (i.e., they discontinued CCT) to the finished water[82, 83, 213, 258]. Thus, the previously formed leaded scale layers began deteriorating and falling into the water[258, 259]. Moreover, without these protective layers, lead-bearing plumbing was in contact with the corrosive Flint River water, and the dissolution of lead from plumbing may have been occurring[95].

Reducing and preventing water lead exposure

The City of Flint reconnected to the less corrosive Detroit water service in October 2015 and enhanced CCT was implemented in December 2015[82, 213, 259]. Replacement of the 29,100 lead and galvanized iron service lines began in February 2016 and is still underway[220]. By once again reducing the corrosivity of the source water and boosting the dose of corrosion inhibitors, the city is reducing WLLs and correcting the damage done to the water infrastructure[259]. In essence, the present-day Flint water has reduced all three elements of lead in the water equation (Figure E.2).

E.5 EFFECTIVENESS OF LEAD IN WATER AVOIDANCE STRATEGIES

When public health officials or residents are concerned about WLLs, four household-level avoidance strategies are commonly recommended: (1) flushing water prior to consumption; (2) using bottled water; (3) installing or using a filter certified under NSF/ANSI 53 to remove lead; and (4) removing lead-bearing plumbing. This section explores WLL reduction efficacy associated with these water avoidance strategies communicated during the Flint Water Crisis and discusses some of the potential financial burdens, maintenance needs, and water conservation implications associated with these remediation strategies (Table E.4 E.5).

E.5.1 Flushing water prior to consumption

Existing public health and utility messages suggest that residents flush water for 30 s to 2 min before consumption[172], as this will remove any stagnant water in contact with lead-bearing plumbing components. In homes where the primary sources of lead are brass fittings or lead-soldered joints in the premise plumbing (Figure E.3(c)), short flushing protocols will often be effective[70, 172, 255]. When partial or full LSLs are present, residents are encouraged to flush a high-volume tap (e.g., bathtub tap) for at least 5 min before flushing the kitchen tap for 1–2 min (Figure E.3(a))[12]. However, even this more extensive flushing protocol is not effective if leaded sediments and unstable scales are present within the system[70, 84, 255, 258, 273]. Thus, flushing protocols must be tailored for specific plumbing configurations and the type of lead release[172]. Moreover, flushing is only effective when consumers follow procedures diligently and frequently, and WLLs can increase rapidly during short periods of stagnation (e.g., 50% dissolved lead after 1 h)[298].

Resident Zero (the Flint resident who uncovered the dimensions of the crisis) was advised by city officials to flush the water for 25 min before consumption due to progressively increasing WLLs in early 2015[313]. However, in-depth testing in April 2015 revealed that all samples collected after a 26-min flushing period (>100 L) still contained lead above 15 µg/L (217–13,200 µg/L)[258]. Moreover, WLLs collected after 20 min of flushing were actually increasing, demonstrating that this advice was ineffective in the midst of the water crisis. During community-wide testing in August 2015, a 3-min flush resulted in a 75% reduction in the first draw 90th percentile WLL (26.8–6.6 µg/L; n = 268), but 47% of residents still had detectable water lead (≤1 µg/L)[259]. After switching back to Detroit water service with enhanced CCT, the city issued a lead advisory, recommending that residents flush the tap water for 5 min before consumption. Water lead samples collected in March 2016 confirmed that 3 min of flushing reduced WLLs for most homes in Flint[259]. Specifically, there was an 86% reduction in the first draw 90th percentile WLL after 3 min of flushing (22.4–3.2 µg/L; n = 156), and 99% of samples were below 15 µg/L. However, 30% of residents still had detectable water lead, and several homes experienced spikes in lead (maximum of 69 µg/L), which were likely linked to the disruption of previously formed leaded scale layers. Thus, sole reliance on flushing as a strategy for reducing WLL exposure appeared to be inconsistently effective during the Flint Water Crisis.

While 3 min of flushing reduced WLLs for most homes in Flint, there were substantial financial burdens and water conservation implications that need to be considered (Table E.4 E.5). Assuming that the water flow rate from a kitchen faucet is 8.3 L/min (2.2 gal/min)[361], flushing for 3 min would have disposed of 24.9 L/flush (6.6 gal/flush). If all 43,404 service connections in Flint flushed twice per day (morning and after work), approximately 2.2 million L (0.6 million gal) would have been flushed daily[16]. Over a 30-day period, this would have amounted to 65 million L (17 million gal). As water service was largely a fixed

cost operation in Flint, flushing practices would have only slightly increased water bills for residents[272]. At $3.30 per 14,195 L (3,740 gal), flushing 3 min twice daily would have cost only $0.01 or $0.35 over 30 days. Therefore, flushing was a low-cost remediation strategy for residents, but was not always effective, and may have increased water and wastewater operation costs and wasted a substantial quantity of water[358].

E.5.2 Consuming bottled water

The USEPA does not regulate bottled water quality, rather the U.S. Food and Drug Administration (FDA) is responsible for the safety and appropriate labeling of bottled water[9]. FDA's bottled water regulations do not pertain to approximately 60–70% of brands, as they are packaged and sold within the same state[240]. There has been considerable debate regarding the health protectiveness of FDA's bottled water standards. But in terms of lead, the FDA has a lower allowable threshold than the USEPA – bottled water must contain less than 5 µg/L, which is a third of the action level[9, 344].

To quantify WLLs in bottled water and potential bottled water lead exposure, our research team analyzed five brands distributed during the Flint Water Crisis (Deer Park, Great Value, Kroger, Member's Mark, and Nestle). All 15 samples contained non-detectable WLLs (<1 µg/L), demonstrating that these brands were safe for lead, and confirming that bottled water is a viable option that can be distributed during a water lead crisis. In addition, other corrosion-related metals (iron, copper, and zinc) were also below detectable levels (<10 µg/L). These brands differed mainly with respect to other water quality factors (e.g., sodium concentrations and water hardness) that can impact aesthetics (more information about bottled water quality and brands can be found at www.nsf.org). While bottled water provides a safe drinking water alternative in terms of lead, there are financial and environmental

implications associated with its use. Moreover, the use of bottled water can impede the recovery of the system, as there will be a limited flow of distributed water with CCT in the premise plumbing and service lines.

Based on our review of popular brands available in grocery stores, a 1-gallon off the shelf container can cost between $0.77 and $1.75, while individual bottles can cost between $0.77 and $8.32 per gallon. Assuming the average person uses 5 L (1.3 gal) daily for all cooking and drinking purposes, a family of four will use 600 L (159 gal) of water over a 30-day period.[21] Using the least expensive bottled water option, this family would spend $122.05 (not including tax) and only save $0.14 on water bill due to the conservation of 20 L/day (assuming $3.30 commodity charge per 14,195 L).[272] As for waste generation, a family could generate 5–40 empty bottles daily and 159–1,200 empty plastic bottles monthly when using gallon and 16.9 ounce bottles, respectively. These estimates are consistent with a CNN profile of the Luster family in Flint.[373] Over 3 days, this family of three used approximately 4.8 gal for cooking, 3.6 gal for drinking water, and 6.9 gal for miscellaneous activities such as washing dishes and brushing teeth. While bottled water provides a safe alternative when tap water is lead-contaminated, this option may not be financially feasible or sustainable for low-income residents. Moreover, due to the inconvenience and expense, residents may use less water – the Luster family used 7 gal/day compared to an average of 100 gal/day.[21]

E.5.3 Using a filter certified to remove lead

There are numerous treatment options available that are certified to remove specific health-related contaminants from drinking water (a consumer tool for identifying water filters certified to reduce lead can be found on the USEPA's website). NSF is one certifying body of water filters. NSF/ANSI 53 certified point-of-use (POU) filters can be a low-cost option

to remediate water lead.[237] POU filters are designed to treat water at a specific outlet, which limits the volume of water needed to be filtered. The activated carbon media sorb dissolved lead and trap particulate lead to reduce WLLs below 10 µg/L for water containing up to 150 µg/L lead.[237] Thus, filters can be an effective remediation strategy if expired filter cartridges are regularly replaced. While POUs are available as tap-mounted, under-the-sink, and pour-through (pitcher filters) from numerous companies, tap-mounted units were recommended and distributed during the Flint Water Crisis.[19]

At the onset of the Flint Water Crisis, there were concerns regarding POU efficacy for homes with WLLs above the NSF/ANSI 53 threshold of 150 µg/L. Our team filtered the worst water lead sample from the home of Resident Zero (sample containing 13,200 µg/L) through an NSF/ANSI 53 certified ZeroWater™ lead filter and observed that 99.85% of the water lead was removed.[208, 258] Although the filtered water was still above the USEPA action level on this extreme 'worst-case' sample, this experiment illustrated that POU filters are effective in dramatically reducing WLLs even under the most extreme conditions. The USEPA conducted additional NSF/ANSI 53 POU testing to examine the efficacy of Brita™ and Pur™ brand filters under more typical 'worst-case' WLLs in Flint (the 90th WLL of the unfiltered sample population was 68µg/L).[19] Based on paired data from 241 Flint homes, POU filters were capable of removing lead in exceedance of 150 µg/L (reduced 4,080 to 0.9 µg/L). Moreover, in most homes, WLLs were reduced to non-reportable levels after filtration (the 90th WLL of the filtered sample population was <0.5 µg/L; high of 1.01 µg/L). Thus, the USEPA data clearly demonstrated that POU filters could effectively reduce the lead in tap water to well below both the action level and the bottled water standard, which is consistent with prior research[91, 92].

Although POU filters were distributed during the Flint Water Crisis at no cost to the residents[19], these devices are also readily available at local stores. Based on our review of

popular brands available, tap-mounted POUs typically cost less than $50, with some models as low as $15. The filter capacity for most units was between 100 and 200 gal (projected to last 3–4 months), and the average cost for a replacement filter cartridge was $10–15. These POU filters cost between $25 and $35, with a replacement filter cartridge costing approximately $10–15. The pitcher style requires more frequent maintenance, as these units are only rated for 25–40 gal (projected to last 1–2 months). Over a 30-day period, the least expensive tap-mounted and pitcher style POU filters would cost only $10–15 in replacement cartridge needs, but this remediation requires an initial purchase of $15–50 for the filtration device. Thus, both POU styles provide a low-cost, effective remediation strategy for residents, but the ease of installation and filter replacement maintenance need to be considered when communicating this strategy.

E.5.4 Removal of partial or full LSLs

Exposure to water lead can be prevented by safely removing sources of lead. In the drinking water infrastructure, LSLs are the most concentrated source of lead and can directly contaminate drinking water. For example, assuming a family of four uses 400 gal of water per day, each foot of ¾" lead pipe contains enough lead to raise every drop of water above the action level for more than 100 years[105]. Thus, replacing the LSL with a non-leaded alternative will greatly reduce water lead exposure.

As previously described, the home of Resident Zero had high WLLs over a 26-min flushing period (>100 L) – the first draw and median WLLs were 2,171 and 1,747 µg/L, respectively[258]. In May 2015, the 25 ft LSL and 192 ft galvanized iron service lines present at the home of Resident Zero were replaced with a non-leaded pipe. The following week, a subsequent lead profile containing 18 sequential 1-L samples was collected at this home. There was a sub-

stantial decrease in WLLs even though there was no corrosion control in the system at that point – the first draw and median WLLs were 2.1 and 1.9 µg/L, respectively. However, there was a spike of 32.4 µg/Lin the 17th liter of water, which is consistent with other literature suggesting short-term incident of high WLLs even after full service line replacements[64, 291]. In addition, this practice still leaves lead sources within the home plumbing, which after the LSL is removed, becomes the source of 100% of the lead in water and can still far exceed the lead action level[333, 335]. Indeed, during the Flint Water Crisis citizen-led sampling events, the worst-case home sampled (WLL of 1,051 µg/L) had no lead pipe, rather the lead was derived from detaching pieces of lead solder[259].

Efforts are currently underway to repair and replace the water infrastructure throughout Flint. It was estimated to take almost a decade to address all infrastructure replacement needs at the previous rate of repair, as only 224 of the 29,100 needed replacements had been completed between February and December 2016[89, 220]. Service line replacement can be a slow process due to its labor-intensive nature (e.g., destruction of streets and sidewalks, removal of complex landscaping, or tree root systems)[16, 89]. The cost of service line replacements can also be cost prohibitive, especially for low-income communities[171]. Additional environmental injustices can arise, as replacement decisions are at the discretion of the property owner, not renters. Through the USEPA funding, LSL and GSL replacements will be done at no cost to Flint residents[22]. However, most cities are not provided this kind of government support. Without state or federal assistance, the cost of a full line replacement ranges from $1,000 to $7,000 per home and may cost more depending on access constraints and other site-specific considerations[187]. When residents are unable to pay for the replacement of the service line on their property, PLSLRs are conducted – a practice that disproportionately impacts low-income residents[171]. Some water utilities are also ‘gifting’ the LSLs, which shift both the ownership and financial burden of LSL replacement to the

customer[169]. Removing the source of lead in drinking water infrastructure is an important step in preventing water lead exposure, but it is a time, labor, and financially intensive process.

E.6 DISCUSSION

When considering potential lead in water exposure and choosing household-level avoidance strategies, it is necessary to understand the protections afforded by regulations and public health guidance. The USEPA's current regulatory framework attempts to account for both water chemistry and infrastructure contributions to lead release by requiring sampling at consumers' taps[344]. However, lead sampling protocols were designed to inform regulatory oversight, not to characterize exposures or public health risks. Moreover, up to 10% of homes sampled during the LCR protocol can contain substantial lead as well as homes not sampled during LCR testing campaigns. Officials are also required to promote flushing on annual consumer confidence reports, and PLSLRs are still required under some non-compliance/exceedance circumstances despite evidence of the short-term-associated risks[171, 172]. While USEPA regulatory officials recognize the limitations of the LCR in protecting all individuals in a city from high water lead exposures, public health officials have long misinterpreted regulatory compliance as public health assurance. The CDC's guidance for investigating the homes of lead poisoned children does not require water lead testing if other sources of lead were found or if the city is meeting the LCR action level[343]. This recommendation has omitted water lead testing in LCR-compliant cities and overlooked a potential route of low-dose chronic lead exposure. These misinterpretations have led to missed opportunities to detect water lead issues and empower people with information on strategies that would allow them to take responsibility for independently addressing potential risks.

The four household water lead avoidance strategies presented in this study reduced WLLs to some degree, but the effectiveness of these avoidance strategies will be site-specific. As explored in Flint, flushing reduced WLLs overall, but the household-level effectiveness was inconsistent. While city officials initially advised residents to flush their taps before using the water, this recommendation was replaced by using bottled water and/or an NSF/ANSI 53 lead filter. This work, along with others[19], have documented that both bottled water and lead filters are strategies that consistently provided safe drinking water. Although replacing the leaded plumbing removes the source of lead, as evident in Flint, this cannot be implemented at the height of a crisis.

For communities concerned about lead in drinking water, building age and knowledge of plumbing materials will help determine the appropriate avoidance strategy, as this information can indicate likely types of leaded infrastructure present. For example, homes with pre-1986 plumbing are more likely to have leaded plumbing sources, including LSLs and lead solder, and may need more intensive interventions like plumbing replacements and avoidance strategies (e.g., bottled water and NSF/ANSI 53 filters). In contrast, lower risk homes, including homes built after 1986 (no LSLs or lead solder) and homes built after 2014 (<0.25% lead in the wetted surface), may not require plumbing replacements, but rather suffice with NSF/ANSI 53 filters. Although flushing is often promoted, it is important to know the presence and location of leaded plumbing materials, as it is not effective when particulate lead and LSLs are present in the system[70, 84, 172, 255]. Lastly, other potential variables impacting the residents and the city, such as financial burden, trash and recycling needs, and water conservation implications are difficult to estimate due to poor record keeping and limited research exploring such costs[358].

E.7 CONCLUSION

There is a critical need for proactive interventions to prevent lead exposure from drinking water instead of relying on reactive regulatory compliance actions that may not be sufficiently protective of public health. In this study, we illustrate the application of an overly simplistic lead in water equation to help understand that worst-case WLLs result from a combination of corrosive water, leaded drinking water infrastructure, and the absence of corrosion controls. Improving any of these conditions can reduce WLLs. The water lead avoidance strategies primarily focus on interventions at the household level to reduce potential water lead, including removal of leaded plumbing, remediation strategies such as flushing and filtration, and complete avoidance by switching to bottled water consumption. The optimal strategy for a given residence will be site-specific and based on a variety of factors and considerations. Thus, engaging with residents and the community will be critical to successful implementation. This work provides public health practitioners, government officials, utility personnel, and concerned residents with science-based information for informed communication, decision-making, and implementation of household-level avoidance strategies for lead from drinking water.

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Appendix F

Supporting Information for Understanding lead in water and avoidance strategies: a United States perspective for informed decision-making

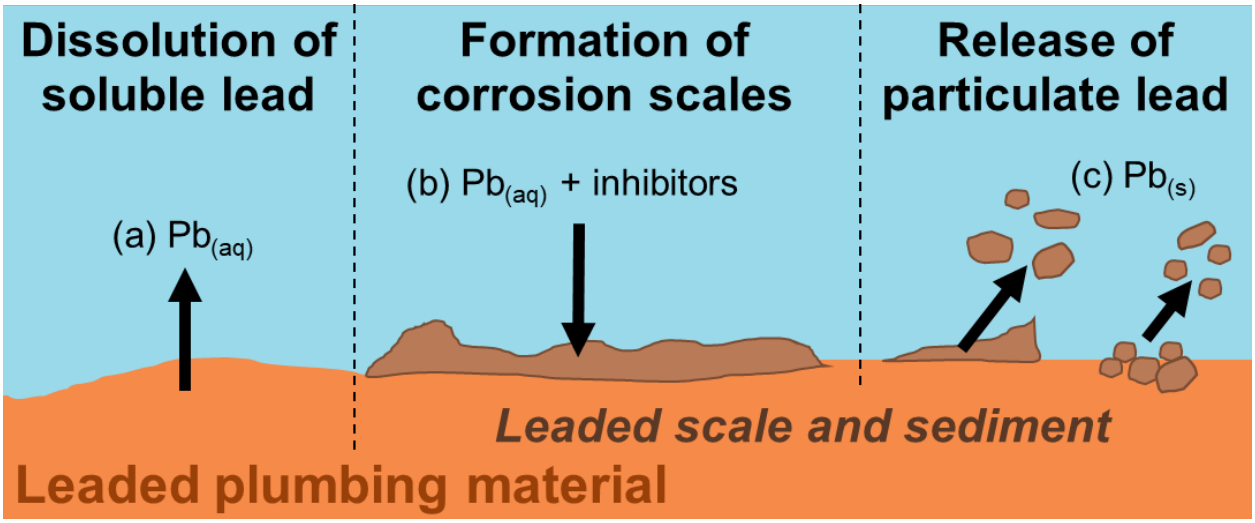


Figure F.1: **Introduction of lead release into drinking water:** a) lead within plumbing components will dissolve into water during periods of stagnation (dissolved lead; $Pb_{(aq)}$); b) corrosion inhibitors to form protective scale layers along plumbing infrastructure; and c) previously formed leaded scale layers can become unstable and detach during periods of water use (particulate lead; $Pb_{(s)}$.



Figure F.2: **Water samples collected at the home of Resident Zero in January 2015**[258]

Table F.1: **Summary of lead-bearing plumbing components used in drinking water infrastructure** (modified from[334]).
[a] Systems with <15 service connections and serving <25 individuals for at least 60 days. Note: private well numbers may be underestimates due to lack of data for private wells nationally[257].
[b] USEPA Lead Ban[4]
[c] USEPA Reduction of Lead in Drinking Water Act[37].
[c] Based on US Census Bureau American Housing Survey unless otherwise noted.
[d] National Survey of Lead Service Line Occurrence[76]
[e] Lead and Copper Rule Revisions: White Paper[20]
[f] Depends on geology and/or driller preference[254]

U.S. home potentially at risk based on year of construction	**Lead-bearing Plumbing Material Potentially Installed**		**Estimated number of U.S. homes (relative percentage) potentially at risk[c]**
Pre-1986	Brass	>8% Pb by weight	90 million (70%)
	Leaded solder	40-50% lead by weight	
	Lead pipe	100% lead by weight	.5->10 million[d,e]
1986-2014[b]	Brass	<8% Pb by weight	128 million (99%)
	Galvanized iron	0.5-1.4% Pb in surface coating	*Public systems*: Service line material used in 52% of utilities surveyed[b]
			Private wells: Unknown[e]
Post-2014[c]	Brass	<0.25% weight average Pb based on wetted surface area	Nearly all homes built since 2014
	Galvanized iron		Unknown
Private wells[a] Pre-1995	Brass well components	>8% Pb by weight	Unknown[f]

Appendix G

Addressing emerging contaminants in well water: a North Carolina perspective on the occurrence and sources of Cr(VI)

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This chapter is planned for submission in mid 2023.

Abstract

Drinking water supplied by private wells poses public health challenges and well users are increasingly concerned about emerging contaminants such as hexavalent chromium (Cr(VI)). In 2018, Hurricane Florence made landfall in North Carolina as a Category 1 hurricane and caused dam failures and flooding of coal ash disposal and reuse sites. Residents surrounding these sites were concerned about potential contamination of their well water with Cr(VI) from coal ash, but there is also widespread naturally-occurring Cr(VI) in the groundwater. To evaluate source attribution techniques we: (1) reviewed literature related to Cr(VI) source tracking and (2) applied these techniques to Cr(VI) data we collected from 1,265 private wells across 22 North Carolina counties to determine if there were differences in sources identified. Almost two thirds of private wells tested (62.0%) exceeded the Cr(VI) public health goal of 0.07 ppb, with concentrations ranging from <0.02-13.9 ppb (median=0.12 ppb). In the literature review, we identified 33 Cr(VI) groundwater tracking techniques from 51 publications and only 5 techniques were used in more than 12 papers. All papers used different combinations of techniques. We applied these techniques to our well sampling data, and inconclusive results were reported for 7 techniques, while three techniques reported potential geogenic and three techniques reported anthropogenic Cr(VI) sources. Specifically looking at coal ash, two techniques did not support coal ash as a primary source and three were inconclusive. Taken holistically, these techniques did not provide definitive insight into Cr(VI) source(s) in the region. This may be due to the fact that these techniques primarily focused on regional scale identification, rather than household-level occurrence. Overall, this study demonstrates the difficulty and complexity in identifying and distinguishing the source(s) responsible for Cr(VI) in well water.

Keywords: Hexavalent chromium; Cr(VI), source tracking, private wells, coal ash

G.1 Introduction

Private wells are the primary source of drinking water for approximately 2.5 million residents (25% of the population) in North Carolina (NC).[96] The 15A NC Administrative Codes define well construction standards and testing requirements for newly constructed and repaired wells, but do not address long-term water quality monitoring.[232] Therefore, well users have sole responsibility for ensuring the safety of their drinking water from private wells. Local health departments have environmental health sections to oversee the enforcement of the Administrative Codes, but the capacity of these programs to assist residents with existing private wells in stewardship activities (e.g., testing, outreach) varies widely.[210, 356] Studies report that well users infrequently monitor and treat their drinking water for health-based contaminants[256, 324] and consumption of contaminated well water has been linked to adverse health effects.[80, 260, 289] Given the challenges well users have with monitoring and remediating well-known contaminants, there are concerns about the well community's ability to address emerging contaminants such as hexavalent chromium (Cr(VI)).[127]

Cr(VI) in groundwater can originate from geologic, non-point sources, and point sources. Geogenic Cr(VI) has been observed in regions containing serpentinite rocks or soils rich in Cr(III), which can be oxidized to Cr(VI) and dissolved in groundwater in the presence of oxidizing agents such as manganese oxide.[244] Agricultural activities have been linked to elevated Cr(VI) levels in groundwater by accelerating natural release of Cr(VI) due to water quality changes associated with increased nitrogen and phosphorus loadings from agricultural runoff.[67, 143] Industries such as mining, leather tanning, and coal burning power plants have also been documented to release waste streams containing elevated Cr(VI), which can then be introduced into groundwater supplies.[43, 86, 349]

In 2014, a pipe break at the Dan River Steam Station in NC released over 100 m^3 of coal

ash into the environment.[153, 234] Coal ash is an end-product of coal burning for power generation, and it contains Cr(VI) along with other toxic metals. Shortly after the spill, the NC legislature passed the 2014 Coal Ash Management Act (CAMA), which was the first comprehensive coal ash management law in the United States.[234] As part of CAMA, private wells within 480 meters (0.3 miles) of coal ash impoundments were required to be tested for a suite of metals, which included a new 0.07 ppb public health goal for Cr(VI).[370] This public health goal corresponds to a one-in-one-million lifetime cancer risk. The NC Department of Environmental Quality (NCDEQ) documented that 68.6% of 341 wells tested surrounding coal ash impoundments contained Cr(VI) above this threshold, with concentrations as high as 22.3 ppb.[228] Because of exceedances of Cr(VI) and other metals, 90.2% of tested wells received Health Risk Evaluations (HREs) from the NC Department of Health and Human Services which stated that the well water was unsafe to use for drinking or cooking.

In September 2018, Hurricane Florence made landfall in NC as a Category 1 hurricane and the historic rainfall caused widespread coastal and inland flooding.[183] The flooding caused dam failures at the Sutton Power Plant near Wilmington, NC, which released an estimated 1,530 m^3 of coal ash into the environment.[351] Several months later, Hurricane Michael also caused flooding across the state. During both storms, other coal ash disposal and reuse sites in the regions were also impacted by flood waters.[188] Residents surrounding these sites were concerned about potential contamination of their well water with Cr(VI) from coal ash. To evaluate Cr(VI) occurrence and identify potential sources in well water, we (1) reviewed literature related to Cr(VI) source tracking to identify published techniques and (2) applied these techniques to data we collected during citizen science campaigns. The overarching purpose of this study was not to validate Cr(VI) source tracking techniques, but rather to demonstrate the difficulty and complexity in identifying and distinguishing the source(s) responsible for Cr(VI) in well water.

G.2 Materials and Methods

G.2.1 Literature review

We identified Cr(VI) source tracking publications using Google Scholar in February 2022. The following search terms were used: ["hexavalent chromium" or "Cr(VI)" or "chromium"] and ["groundwater" or "polluted groundwater" or "coal ash"]. We reviewed papers with titles referencing Cr(VI) in groundwater, except when papers focused on Cr(VI) removal via treatment technologies. Overall, 77 papers were identified. Papers discussing the presence of Cr(VI) rather than potential sources (n=13) and papers focusing on attenuation of plumes from previously identified sources (n=13) were removed. In total, 51 papers were included in this study.

We identified 33 techniques used (Supporting Information Table H.1) to identify Cr(VI) sources. We grouped these techniques into 7 general approaches (1) "geologic setting" addressing the potential of aquifer materials to release Cr(VI); (2) "spatial analysis" exploring proximity to Cr(VI) sources; (3) "correlations" between Cr(VI) and water quality parameters; (4) use of "multivariate analysis"; (5) comparison to water quality "thresholds"; (6) evaluation of "isotope ratios"; and (7) use of "predictive modeling" software. Specifically, the "geologic setting" approach represents techniques that used data related to aquifer materials (e.g., rock types) from published datasets, collection of sediment samples, or data from leaching tests. The "spatial analysis" approach represents techniques that used relative distances or locations to known Cr(VI) sources. The "correlation" approach represents techniques that identified associations between water quality parameters. The "multivariate analysis" approach included multivariate statistical approaches (e.g., ordination analysis, cluster analysis). We defined the "threshold" approach as techniques that relied on estab-

lished thresholds to differentiate between Cr(VI) sources. The "isotope ratio" approach represents techniques that evaluated differences in isotopic ratios between geogenic and anthropogenic Cr(VI) sources. The "predictive modeling" approach included any techniques that developed predictive models based on potential sources to determine relationships between Cr(VI) and/or other water quality parameters.

G.2.2 Citizen science sampling campaign

Between October 2018 and November 2019, we hosted well water testing clinics in counties with flood-impacted coal ash impoundments, landfills, and structural fills ("coal ash sites"). In New Hanover County, coal ash stored in an impoundment was released into the environment during the dam failure at the Sutton Power Plant.[351] In Iredell County, concerns over coal ash from the Marshall Steam Station[234] were heightened when coal ash structural fill at a construction site adjacent to a school was exposed by erosion during the heavy rainfall.[188] Additionally, there was widespread flooding reported in Robeson, Chatham, and Northampton counties, which are home to the Weatherspoon Power Plant, the Brickhaven Mine and Cape Fear Power Plant, and the Kapstone landfill, respectively.[234] We hosted sampling clinics in Chatham (240 samples), Iredell (787 samples), New Hanover (82 samples), Northampton (92 samples), and Robeson counties (85 samples), but allowed residents in neighboring counties to participate (Figure H.3).

We advertised our free testing opportunities through local media outlets and community organizations (e.g., health departments, non-profits, NC Cooperative Extension programs). Residents were provided sampling kits that contained instructions, a household survey, and sampling bottles. In New Hanover and Robeson counties, participants flushed a working tap for 5 minutes before collecting a 250 mL bottle for "flushed" metal analysis and 125 mL

bottle for Cr(VI) analysis. In Iredell, Chatham, and Northampton counties, participants collected a 250 mL bottle after 6+ hours of stagnation for "first draw" metal analysis and then collected the two flushed samples as previously described. After sample collection, participants were asked to complete a household survey, which included information about well type and depth and treatment devices installed. Participation in this campaign was voluntary and all procedures were approved by Virginia Tech Institutional Review Board (#16-918 and #19-149).

We measured pH and conductivity immediately after samples were received from residents per EPA Methods 150.3 and 120.1,[72] and transferred a 10 mL aliquot from the flushed metals bottle for anions analysis using an ion chromatography column (Dionex ICS-1600) per method 300.0.[252] Sampling kits that were shipped instead of collected during clinics did not receive pH or anion analysis (n=92). First draw and flushed metal samples were acidified with 2% nitric acid and digested for 16+ hours before analysis using inductively coupled plasma mass spectrometry (Thermo Scientific iCAP RQ ICP−MS) per method 3125 B.[72] Ammonium based liquid preservative was added to the Cr(VI) bottle per EPA method 218.7 before kits were distributed to residents, and samples were analyzed using a modified ion chromatography column (Dionex ICS-1000).[371] Detection limits were 0.1 ppb for most metals, with the exception of iron (5.0 ppb), copper, zinc, and selenium (1.0 ppb), strontium (0.5 ppb), and Cr(VI) (0.02 ppb).

G.2.3 Well water citizen science campaigns

Overall, we included 237 samples from Chatham, 780 samples from Iredell, 80 samples from New Hanover, 92 samples from Northampton, and 76 samples from Robeson sampling clinics in our analysis. Samples collected from non-private well sources (n=6), duplicate samples

from the same well (n=5) and tap samples when wellhead samples were also collected (n=10) were removed. Addresses were not provided for 8 wells and these systems were not included in analysis requiring well locations. To determine well locations, we geocoded sampling addresses using the ArcGIS World geocoding service. We matched well locations with primary aquifer designations (Figure H.3) using the USGS Ground Water Atlas.[6] We collected samples from four primary aquifer systems: Castle Hayne (n=60), Northeastern Atlantic Coastal Plain (n=149; "Northeastern Atlantic"), Piedmont and Blue Ridge crystalline-rock (n=1,015; "Piedmont and Blue Ridge"), and Surficial (n=28) aquifers. Aquifers could not be identified for 13 wells because of missing address and/or well depth.

G.2.4 Source tracking analysis

Source tracking analyses were conducted in R (version 4.1.0) and ESRI ArcGIS (version 10.6.1). Details about the source tracking techniques we applied are presented in Section G.3.1. In brief, we evaluated water quality characteristics and exceedances using summary statistics. We used the Kruskal-Wallis test to determine differences between Cr(VI) concentrations between aquifer systems. For the remaining analyses, we only used data from the Piedmont and Blue Ridge aquifer because of the presence of geogenic Cr(VI), large sample size (n=1,015), and high occurrence of Cr(VI) (i.e., 72.0% of samples above 0.07 ppb). We used Spearman correlations to evaluate relationships between Cr(VI) and other contaminants and distances to known coal ash sites and we measured distances between well locations and the nearest coal ash site. We used kriging to display the spatial distribution of interpolated Cr(VI) values, with manual breaks of Cr(VI) concentrations of below detection (0.02 ppb), between detection and the public health goal (0.07 ppb), and then incrementally up to the NC 2L total Cr standard (10 ppb). We downloaded land use data from the Multi-Resolution Land Characteristics Consortium[3] to determine the land use type for each

sampled well. We categorized land use as either developed ("Developed, High Intensity", "Developed, Medium Intensity", "Developed, Low Intensity", "Developed, Open Space"), agriculture ("Hay/Pasture", "Cultivated Crops"), or undeveloped (all remaining categories). Principal component analysis (PCA) and K-means clustering were conducted for all water quality data. For both analyses, parameters with greater than 30% of samples below detection were removed in accordance with previous studies (12 parameters removed).[350] For the PCA, data were scaled, and results were evaluated using the FactoMineR package. For cluster analysis, data were scaled prior to cluster formation using the kmeans package. We conducted a Tobit regression (i.e., censored linear regression model), with a total percentage of censored values of 10.8%. We evaluated the relationship between Cr(VI) and pH and conductivity in the Piedmont and Blue Ridge as well as across all aquifers.

G.3 Results

G.3.1 Cr(VI) source tracking review

In total, 51 papers were reviewed to characterize source tracking techniques used to identify Cr(VI) source(s) in groundwater. We observed that Cr(VI) was generally documented in the literature to originate from one of three sources – (1) geogenic sources primarily from mafic or ultramafic rocks, (2) anthropogenic sources from coal ash or other industrial waste, and (3) effects of agricultural activity on groundwater chemistry causing accelerated release (Table H.2). Most papers (32 papers) considered two or more potential Cr(VI) sources. Geogenic Cr(VI) was among the sources considered in most papers (44 papers) with industrial contamination considered in 28 papers and agricultural activity in 16 papers. Industrial contamination was rarely associated with coal ash, as only 6 papers considered coal ash in

their source analysis. Among these 6 studies, one determined that Cr(VI) was attributed to coal ash,[175] two determined Cr(VI) was not associated with coal ash,[350, 352] and three were unable to differentiate coal ash from other Cr(VI) sources.[170, 287, 357]

We identified 33 techniques that were used to determine Cr(VI) source(s) (Table H.1). Analysis of published data regarding rock types (40 papers) and mapping of Cr(VI) values relative to potential sources (40 papers) were the most commonly used techniques. Construction of ternary diagrams to evaluate groundwater type (22 papers), analysis of sediment or aquifer material samples (19 papers), and analysis of the correlation between Cr and Cr(VI) were relatively common, but no other single technique was used by more than 12 papers (23%). The number of techniques used per paper varied from 2 to 9, with 5 techniques being most commonly used (Figure H.4). Interestingly, no papers used the same combination of techniques, as each developed their own combination. All 6 papers that investigated the contribution of Cr(VI) from coal ash mapped locations of Cr(VI) concentrations and analyzed published rock type data, with three papers also creating ternary diagrams to classify groundwater types. Other methods were less frequently used.

We combined the 33 techniques into 7 general approaches (Table H.1). Geologic setting (45 papers), spatial analysis (41 papers), thresholds (33 papers), and correlations (30 papers) approaches were used in more than half of papers. Nearly all studies (50 papers) used more than one general approach in their analysis, and we identified 11 combinations of approaches used by more than one paper (Figure H.5). However, there was little consistency in which combination of approaches was used, with the most common combination only used in 7 papers. For the identification of coal ash, all 6 studies used geologic setting and spatial analysis, none used a predictive modeling approach, and 2-3 studies used techniques in the other 4 approaches.

The scale at which Cr(VI) source tracking techniques were applied often covered a regional area, as studies most commonly (27 papers) analyzed samples from multiple aquifer systems to determine probable Cr(VI) sources in the region. Other studies (20 papers) focused on a single aquifer system, although these often spanned large areas (100-400 km^2) and contained multiple geologic formations. Among these 47 studies, samples were commonly collected from private or irrigation wells, but sometimes also included samples from springs, surface water, or monitoring wells. The remaining four studies focused on a specific site to determine localized Cr(VI) sources, and these studies sampled monitoring wells.

The various techniques and approaches offered different methodologies for source tracking depending on available data and the Cr(VI) sources considered. However, we observed that no combination of techniques or approaches was consistently applied, highlighting that there is no standardized approach for determining Cr(VI) sources in groundwater. In addition, most of the techniques did not report sources of Cr(VI) impacting a single well or home, but instead focused more on general Cr(VI) sources across one or multiple aquifer systems.

Geologic setting

These techniques aimed to determine background levels of geogenic Cr(VI) within an aquifer using data characterizing the aquifer material. With this information, authors were able to estimate background levels and compare concentrations to observed Cr(VI) measurements to determine the potential contribution from anthropogenic sources (e.g., Moraetis 2012,[222] Panagiotakis 2015[245]). However, these techniques were not able to identify the anthropogenic source(s), creating the need for additional approaches if source identification was needed. Of the 45 studies using this approach, most (n=23) conducted sampling across multiple aquifer systems for regional-scale Cr(VI) source identification while others (n=18) focused on one aquifer system but still aimed to identify regional-scale Cr(VI) sources. Only

4 studies focused on a specific site to identify localized Cr(VI) sources (e.g., Bertolo 2011[52]).

We identified three geologic setting techniques: (1) analyzed published data regarding rock types in region, (2) analyzed Cr content of samples of aquifer material, and (3) performed leaching tests to determine potential of aquifer materials to release Cr(VI). We used two techniques in our study, as we had access to data on rock types in the region.[6] The other two techniques required sediment and leaching data which we did not collect.

Spatial analysis

These techniques determined if Cr(VI) was at higher concentrations in closer proximity to known sources or distributed uniformly through the study area (e.g., Vasileiou 2019[350]). However, this approach could not identify non-point sources of Cr(VI) or distinguish the contribution of Cr(VI) from multiple sources (e.g. Hausladen 2018[143]). Of the 41 studies using this approach, 22 conducted sampling across multiple aquifer systems while 17 focused on one aquifer system to identify regional Cr(VI) sources. The remaining studies (n=2) focused on specific sites for localized Cr(VI) source identification (e.g., Dermatas 2016[86]).

We identified three spatial analysis techniques: (1) mapped Cr(VI) values relative to potential sources, (2) evaluated distance between measured Cr(VI) value and potential sources, and (3) mapped Cr(VI) values relative to published land-use data in the region. We used all three techniques in our study, as we had spatial locations of each sampled well, land-use data,[3] and had the locations of coal ash sites.

Correlations

These techniques included correlations with elements commonly found in aquifer materials (e.g. silica, vanadium, selenium, barium) which were used to indicate geogenic Cr(VI)

(e.g. Armienta 1993[43]), while correlations with elements associated with human activities (e.g. nitrate, ammonia, chloride) were used to indicate anthropogenic sources (e.g. Manning 2015[211]). In addition, many studies (e.g. Vengosh 2016[352]) identified a strong (ρ>0.90) correlation between Cr(VI) and total Cr, confirming that Cr(VI) is the predominant species of dissolved Cr in groundwater. Papers within the same technique did not always use the same combinations of elements; i.e. Panagiotakis 2015[245], considered correlations between Cr(VI) and Ni as an indicator of natural sources while Paternoster 2021[250] cited correlations between Cr(VI) and Si, Mg, and HCO_3^-. No paper required significant correlations between all parameters explored to draw conclusions about potential sources. Therefore, we evaluated correlations observed between parameters to determine sources rather than the combination used in each study. Of the 30 studies using this approach, nearly all focused on regional Cr(VI) source identification, either within one aquifer system (n=11) or across multiple systems (n=18). Only one paper used the correlations approach to evaluate Cr(VI) sources at a specific site.

We identified 10 correlation techniques: evaluating correlations between (1) total Cr and Cr(VI), (2) all measured parameters including Cr(VI), (3) Cr(VI) and Si, Ca, Ni, Fe, Mg, HCO_3^-, V, Se, U, Ba, and Co to determine natural origins, (4) Cr(VI) and Mn to determine natural oxidation of Cr(III) by Mn oxides, (5) Cr(VI) and V, Se, U, and Ba to determine similar natural oxidation/reduction pathways, (6) Cr(VI) and Na^+ and Ca^{2+} to determine natural ion exchange in aquifers resulting in Cr(VI) desorption, (7) Cr(VI) and NO_3^-, NH_3, DDE, or P to determine agricultural sources, (8) Cr(VI) and chlorinated solvents (i.e. DCE) to determine industrial contamination, (9) Cr(VI) and Mg, Ni, and B within plume, and (10) Cr(VI) and Cl^-, SO_4^{2-}, HCO_3^-, Ca^{2+} to determine natural Cr(VI) as opposed to anthropogenic ions. We used 8 of these techniques in our study, as parameters mentioned by these techniques were collected in our study. The remaining techniques required data that were

not collected in our study.

Multivariate analysis

These techniques include analyses to examine both gradients (i.e., via ordination analyses) and discontinuities (i.e., via cluster analyses) in datasets. Specifically, studies used combinations of these techniques to develop groups of sampled wells with similar water quality results and evaluated similarities within groups (e.g. Yerima Kwaya 2019[369]). All papers either used a standalone PCA (i.e. Sakizadeh 2019[287]), a standalone factor analysis (FA) (i.e. Stamatis 2011[317]), or used a PCA to develop a FA (i.e. Tamma Rao 2013[326]). No paper used both FA and PCA for results analysis. Of the 13 studies using this approach, all focused on regional Cr(VI) source identification, either within one aquifer system (n=6) or across multiple systems (n=7). This approach was not applied for site-specific Cr(VI) source identification.

We identified 3 techniques: (1) performed PCA of water quality parameters, (2) performed FA using physical and chemical parameters of water samples, and (3) performed cluster analysis of water quality parameters. We used two techniques in our study, which both used available water quality data. A FA was not conducted due to its redundancy with PCA results. It is important to note that the goal of our analysis was to use published techniques. While there are other techniques that may be appropriate, this was not within the scope of our study.

Thresholds

Eight studies used thresholds based on values observed in previous cases of interactions between water and mafic rocks to identify geogenic Cr(VI) (e.g. Charoula 2021[67]). Other

studies developed thresholds for coal ash contamination[352] or for differentiating between natural and anthropogenic contamination.[88, 326] However, it is important to note that thresholds can be specific to study sites due to the natural variation between aquifers and pollution sources. Of the 33 studies using this approach, 16 conducted sampling across multiple aquifer systems while 14 focused on one aquifer system to identify regional Cr(VI) sources. Remaining studies (n=3) focused on specific sites for localized Cr(VI) source identification.

We identified 7 threshold techniques comparing: (1) water quality results to established coal ash contamination thresholds for B and Sr, and B/Cr and Sr/Ca ratios, (2) pH, DO, water quality, and redox potential from different wells to establish relative water age, (3) Ca^{2+}/HCO_3^- ratio between wells to determine relative depth of groundwater, (4) ternary diagrams of major ions of differing groundwater types, (5) HCO_3^-/Mg, Ca/Mg, SiO_2/HCO_3^-, Ca/Si and Mg/Si ratios with those observed in groundwater containing known geogenic Cr(VI), (6) Cr(VI) results to rough geogenic thresholds of 50-100 ppb, and (7) distribution of observed data to a normal distribution. We used 3 techniques in our study as we collected data on Cr(VI), B, Sr, Ca, Mg, and Si concentrations. We were not able to use the remaining techniques because the data needed were not collected during our campaign.

Isotope ratios

Five studies used a Cr isotope ratio ($^{53}Cr/^{52}Cr$) to determine the extent of Cr(VI) reduction and track the remediation of known plumes,[151, 354] understand natural reduction processes,[104, 175] and differentiate between the two.[168] One paper compared a Sr isotope ratio ($^{87}Sr/^{86}Sr$) in waters associated with coal ash to natural waters to determine similarities and differences. Of the 6 studies, all focused on regional Cr(VI) source identification, either within one aquifer system (n=5) or across multiple systems (n=1). This

approach was not applied for site-specific Cr(VI) source identification.

We identified two isotope techniques: (1) determining $^{53}Cr/^{52}Cr$ ratio to examine the degree of reduction of Cr(VI) and (2) comparing $^{87}Sr/^{86}Sr$ ratios to differentiate natural and anthropogenic contamination. Neither isotope ratio technique was used in our study because we did not collect isotope data.

Predictive modeling

These techniques used process-based models to understand chemical kinetics and predict contaminant concentrations, through both chemical modeling of geogenic Cr(VI) release from rocks[42, 87] and groundwater modeling to simulate Cr(VI) flow and variability.[222] Other studies used water quality data to make statistical estimations of contaminant concentrations, through regression modeling of water quality data[79] or geostatistical modeling to identify contamination hotspots.[168] Various studies also used chemical reaction modeling software (e.g. Rayleigh distillation model[104] or MINTEQA2[52]) to estimate the solubility and speciation of chromium in drinking water. Of the 17 studies using this approach, 7 conducted sampling across multiple aquifer systems, 8 focused on one aquifer system to identify regional Cr(VI) sources, and two focused on a specific site for localized Cr(VI) source identification.

We identified five modeling techniques: (1) modeling aquifer materials and solute conditions to estimate potential for geogenic Cr(VI) release, (2) constructing regression models from parameters that can be measured in the field (i.e. pH, TDS, DO) to estimate probability of high geogenic Cr(VI), (3) developing geostatistical modeling to identify clusters of high concentrations of water quality parameters, (4) modeling chemical reactions occurring in drinking water to estimate solubility and speciation of Cr(VI), and (5) conducting ground-

water modeling to simulate Cr(VI) variability due to observed results. Such models required a variety of data types for model inputs to accurately model the system, which makes them difficult to apply when potential sources and transport are not already identified. We used one technique in our study as we had data for pH, conductivity, and primary aquifer located in for each well. The remaining techniques were not used because the data needed were not collected in our campaign.

G.3.2 Application of source tracking techniques

Almost two thirds of private wells tested (62.0%) exceeded the Cr(VI) public health goal of 0.07 ppb, with concentrations ranging from <0.02-13.9 ppb (median=0.12 ppb; Table H.1). This occurrence of Cr(VI) is consistent with previous studies in NC private wells, which observed similar concentrations in the Piedmont and Blue Ridge Aquifer.[78, 79, 352] When reported (n=943), 89.1% of users reported having drilled wells, and less reported use of dug or bored (10.4%) or sand point wells (0.5%). The median Cr(VI) concentration was higher ($p<0.01$) in drilled wells (0.14 ppb) compared to dug/bored (0.07 ppb) and sand point wells (<0.02 ppb). Depth was reported in 59.0% of wells, and wells deeper than 100 ft had a higher ($p<0.01$) median Cr(VI) concentration (0.17 ppb) than wells with depths of 51-100 ft (0.08 ppb) and 0-50 ft (<0.02 ppb).

To evaluate the agreement of the various source tracking techniques in identifying Cr(VI) sources in drinking water from private wells, we applied 18 of 33 techniques to the results of our well sampling campaign to explore their agreement with Cr(VI) sources identified in our study.

Geologic setting

Cr(VI) concentrations in the Piedmont and Blue Ridge aquifer (n=1,015) were significantly higher than other aquifers (Kruskal-Wallis Test, p<0.01; Figure G.1), with a median concentration of 0.26 ppb compared to <0.02 ppb in the Castle Hayne (n=64), Northern Atlantic (n=150), and Surficial (n=29) aquifers. In addition, Cr(VI) concentrations exceeded the public health goal in 72% of wells in the Piedmont and Blue Ridge compared to 14%-22% of wells in the other aquifers. Within the Piedmont and Blue Ridge aquifer, more than two thirds of wells (69.0%) were constructed within formations that contained mafic rocks such as biotite.[6] All wells within the Castle Hayne (carbonate-rock), Northern Atlantic (semi-consolidated sand), and Surficial (unconsolidated sand and gravel) aquifer contained materials that are unlikely to contain mafic rocks.[6] Median Cr(VI) concentrations were significantly higher (p<0.01) in formations with mafic rocks (0.34 ppb) than in other formations (0.13 ppb) within the Piedmont and Blue Ridge aquifer. Within these formations, there was no significant difference in Cr(VI) (p=0.42) between drilled wells (n=504, median=0.28 ppb) and dug/bored wells (n=39, median=0.41 ppb). These findings suggest that groundwater in the Piedmont and Blue Ridge region can contain geogenic Cr(VI). This is consistent with Coyte 2020[79], who documented that 66% of sampled wells in the Piedmont and Blue Ridge exceeded the 0.07 ppb public health goal (median=0.63 ppb) compared to 18% in other aquifers (median <0.5 ppb). Similarly, Vengosh 2016[352] recorded Cr(VI) ranges of 0.01 to 22.9 ppb, with 90% of wells above 0.01 ppb within the Piedmont and Blue Ridge aquifer.

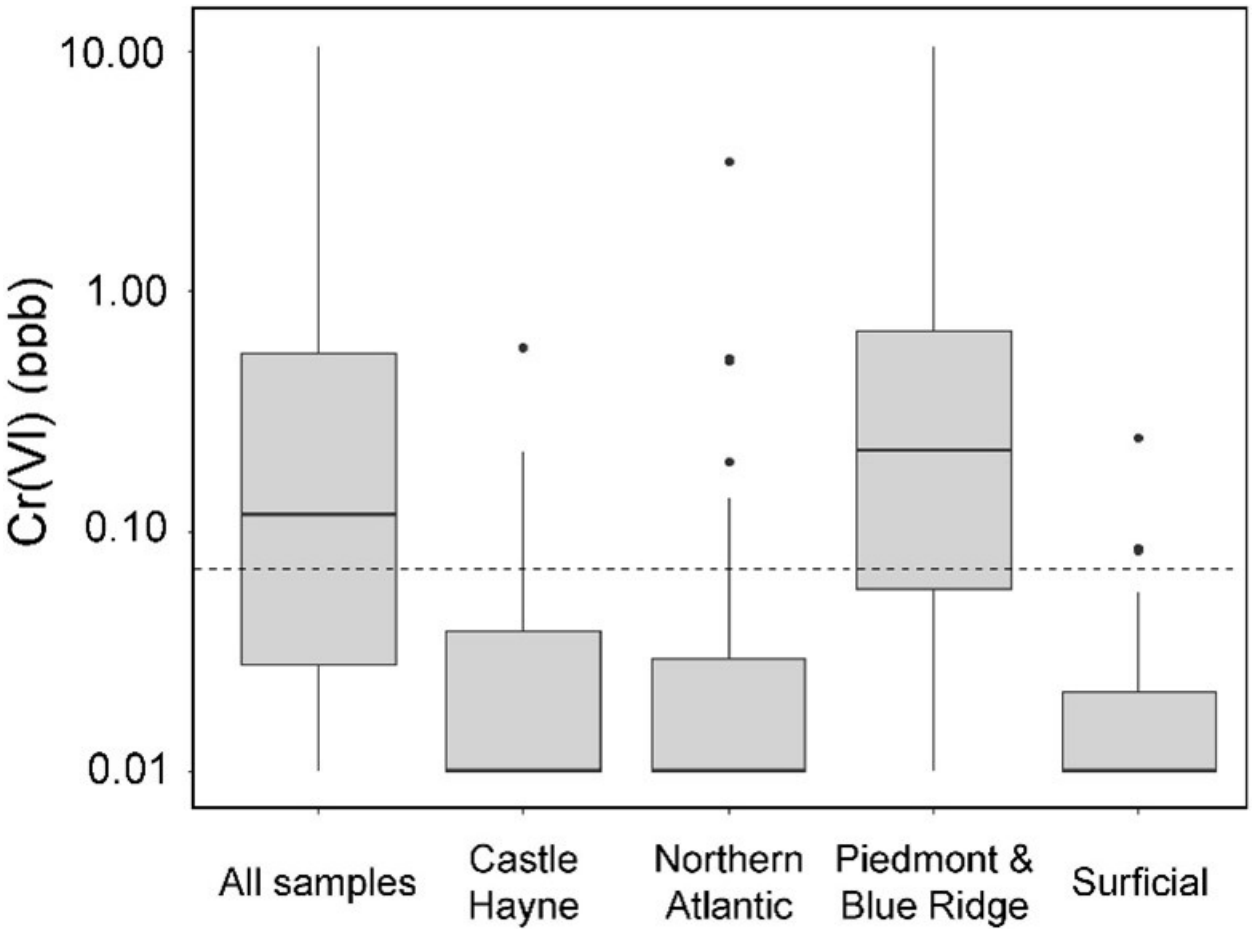


Figure G.1: **Cr(VI) concentrations by aquifer system.**
All data below the detection limit of 0.02 ppb were set to 0.01 ppb. The dashed line indicates the 0.07 ppb NC public health goal.

Spatial analysis

When mapping Cr(VI) concentrations relative to coal ash sites, concentrations were not elevated near known sites. In the Iredell clinic, two areas with interpolated Cr(VI) values of 5.0-10.0 ppb were located in the northeast and western parts of the study area. Interpolated Cr(VI) concentrations around the Marshall Steam Station were 0.07-2.5 ppb (Figure H.6). In the Chatham clinic, there were no areas with interpolated Cr(VI) values of 5.0-10.0 ppb and the coal ash sites were located in an area with interpolated Cr(VI) concentrations of 0.02-0.07

ppb (Figure H.6). These results suggest known coal ash sites were not the primary Cr(VI) source of well water in these regions. However, this approach was not able to distinguish or quantify Cr(VI) contributions from multiple sources.

The private wells in our study were located 1.2-31.7 miles from a coal ash site and all were outside the 480 m (0.3-mile) radius outlined in CAMA (Figure G.2). Cr(VI) concentrations were not correlated with distance to a coal ash site (ρ=-0.21), and Cr(VI) concentrations were above 0.07 ppb up to 49 km (31 miles) away. These results were in keeping with Vengosh 2016,[352] who did not observe a correlation between Cr(VI) and distance to known coal ash sites (r=0.09), and documented Cr concentrations up to 34 ppb more than 50 km (32 miles) from the nearest site.

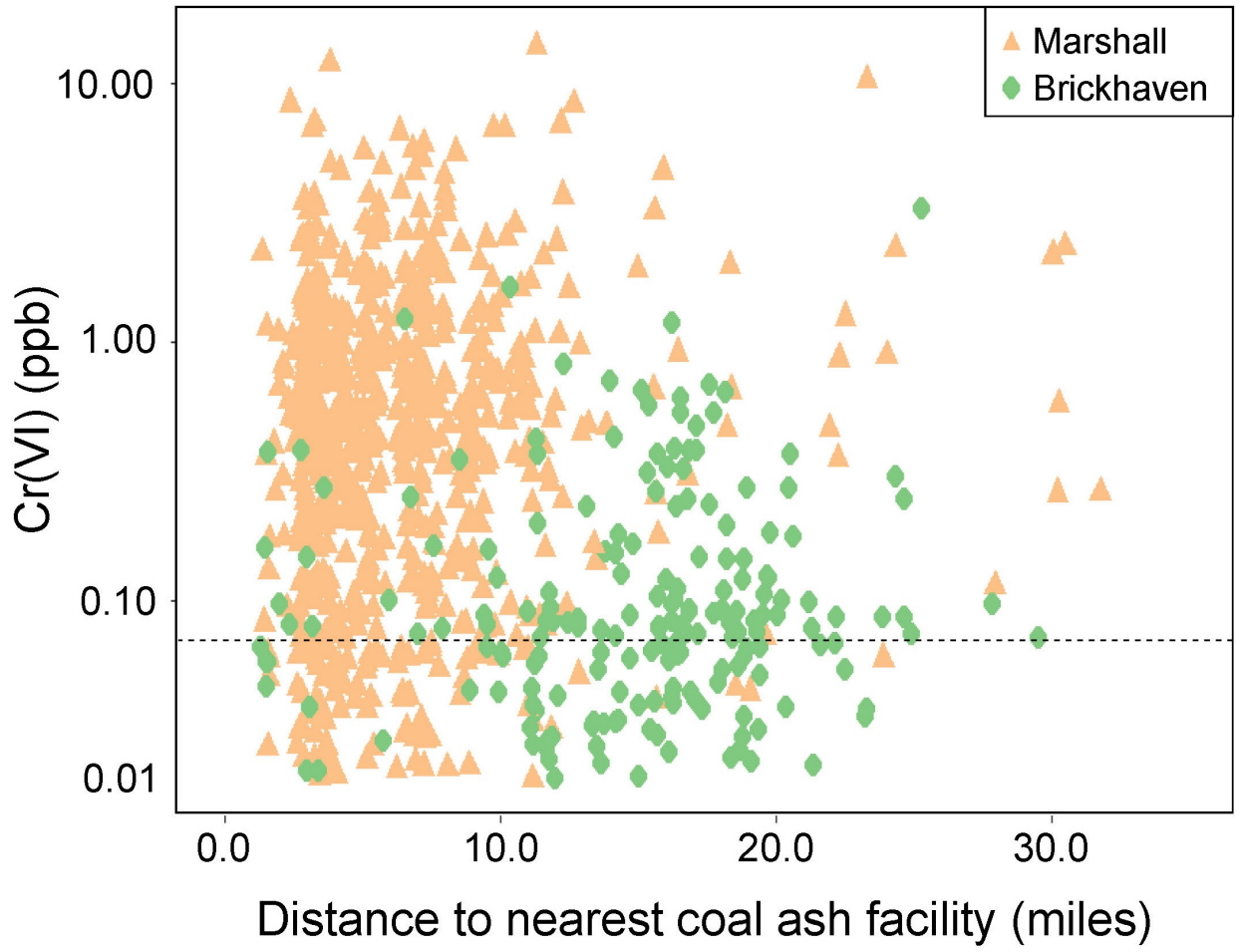


Figure G.2: **Comparison of Cr(VI) concentrations** measured during our study and distance to the Marshall Steam Station (MSS) in Iredell County (orange) or the Brickhaven Mine and Cape Fear Plant in Chatham County (green). The dotted line indicates the NC public health goal of 0.07 ppb.

Most wells in our study (65.2%) were within developed areas, 8.7% were within agricultural areas, and 25.6% were within undeveloped areas. Cr(VI) concentrations were different (Wilcox Test, $p<0.05$) between land use categories, with the highest median value (0.40 ppb) occurring in developed areas, followed by agricultural (0.35 ppb), and undeveloped areas (0.18 ppb). Kumar Tiwari 2019[331] observed the highest Cr(VI) concentrations in the industrial zone in their study, concluding that industrial sources of Cr(VI) were an impor-

tant source. Other studies([217, 340] have found the highest Cr(VI) levels in the agricultural zone, suggesting farming activity is an important contributor. Based on these studies, our results suggest potential anthropogenic contributions in this region but do not distinguish sources.

Correlations

Correlations between Cr(VI) and 16 of the 20 of elements used in the literature were analyzed (Table H.2). Seven elements (Si, Fe, V, Se, Co, Mn, Na) were associated with natural Cr(VI) sources, 5 (NO_3^-, B, Cl, SO_4^{2-}, P) were associated with anthropogenic sources, and 3 (Mg, Ni, Ca) were associated with both. Cr(VI) was correlated with total Cr (ρ=0.68) (Figure H.7), which is consistent with prior studies highlighting that Cr(VI) is the predominant species of dissolved Cr in groundwater and that total Cr may be predictive of Cr(VI) concentrations.[79, 352] Cr(VI) was also correlated with V (ρ=0.52), Na (ρ=-0.44), SO_4^{2-} (ρ=-0.42), Mn (ρ=-0.35), and B (ρ=-0.33). Weak correlations were observed between Cr(VI) and Si (ρ=0.25), NO_3^- (ρ=0.23), and Cl (ρ=-0.26). No correlations were observed between Cr(VI) and Ca, Co, Fe, Mg, Ni, P, or Se.

Similar correlations between Cr(VI) and V (ρ=0.42 and 0.40) were observed in prior studies,[143, 165] where the authors suggested that this relationship indicated geogenic Cr(VI) sources. Additionally, Cr(VI) correlations with SO_4^{2-} and Cl were in agreement with prior studies (no ρ provided),[67, 189] stating that water lacking in Cl and SO_4^{2-} has not been altered by anthropogenic activities and therefore indicated geogenic Cr(VI) sources. However, other studies documented strong correlations between Cr(VI) and Si (ρ=0.63 to 0.92) from geogenic contributions,[250, 350] while we only observed a weak correlation in our study. In addition, moderate correlations between Cr(VI) and NO_3^- (ρ=0.39 to 0.47) have been linked to agricultural sources,[54, 67, 143] while we only observed a weak correlation in our study.

Although we observed multiple correlations outlined in prior literature, there were numerous instances when we did not observe documented correlations. For geogenic Cr(VI) sources, previous studies[136, 140, 165, 175, 245, 250, 350, 357] have identified correlations between Cr(VI) and Ca (negative correlation), Mg (ρ=0.31 to 0.66), Mn (no ρ provided), Na (R^2=0.15), Ni (ρ=0.42 to 0.64), and Se (ρ=0.29). For anthropogenic sources, one study[86] identified correlations between Cr(VI) and Mg (ρ=0.33), Ni (ρ=0.58), and B (ρ=0.65), which were not observed in our study. The lack of these correlations does not provide evidence for geogenic release or anthropogenic sources in the area, rendering these techniques inconclusive.

We developed a correlation matrix of all measured parameters. Correlations were observed between Ca and Mg (ρ=0.79), Ca and Ni (ρ=0.44), Mn and V (ρ=-0.40), and B and SO_4^{2-} (ρ=0.40). One study[161] observed a correlation between Ca and Mg (r=0.80), stating these parameters share a common source. No other paper made conclusions about Cr(VI) sources based on correlations we observed in our study. Other studies highlighted correlations between Ca and SO_4^{2-}, Mg, Si, and Fe, or Ni and Mg to suggest geogenic sources. Overall, the lack of expected correlations observed in our study is inconclusive to determining the presence of geogenic or anthropogenic sources.

Multivariate analysis

We used a combination of PCA and cluster analyses to examine gradients and discontinuities in our dataset. No principal component explained more than 17.8% of variance and 9 components were retained with eigenvalues greater than 1 (Figure H.8). The first principal component included strong (>0.6) positive loadings for conductivity, Cl, Mg, Ca, Ti, Mn, and Br and a weak negative (0.2) loading for Cr(VI) (Figure H.9). The second component included moderate (>0.4) positive loadings for Si, P, Br, and I, and moderate (<-0.4) negative loadings for pH, SO_4^{2-}, W, and Mo. The third component included moderate positive

loadings for Ca, Ti, and Sr, and a strong negative loading for Na. Loadings observed in these components were not consistent with components previously used in literature to identify potential Cr(VI) sources. For example, Vasileiou 2019[350] identified pairings of Ca and HCO_3^- and Cr, Cr(VI), Si, and Mg to suggest the influence of interactions with local geology as a common source. Other papers[247, 350] identified pairings of Cr(VI) with NO_3^-, Si, and P to indicate agricultural or other anthropogenic sources. Therefore, this technique was inconclusive for source identification.

In the k-means clustering, no drastic change in slope was observed (Figure H.10), so we selected 4 groups due to the gradual change in slope between 4 and 5 groups. The resulting grouping produced Groups 1-4 (Figure H.11) which consisted of 150, 62, 501, and 19 private wells. Cr(VI) values in Group 2 were higher (Wilcox Test, $p<0.05$) than other groups with a median value of 0.55 ppb compared to 0.06-0.08 ppb (Table H.3). In addition, Group 2 had higher P and NO_3^- concentrations than other groups (median P concentrations of 41.1 ppb compared to 5.6-33.1 ppm and NO_3^- concentrations of 0.88 ppm compared to 0.12-0.24 ppm). These results are in keeping with Vasileiou 2019[350] who previously found one group of wells with higher Cr(VI), NO_3^-, and P stating that this indicated potential effects of agricultural activity. Therefore, this technique would suggest agriculture as a source of Cr(VI) in this region.

Thresholds

Thresholds for detecting coal ash contamination were developed in NC by Vengosh 2016.[352] The authors concluded that lack of exceedance of B, B/Cr, and Sr/Ca ratios indicated a lack of influence by coal ash. We observed B concentrations above 100 ppb in only 0.2% of wells, and exceedance of the B/Cr ratio threshold of 200 in only 0.7% (Table H.4). However, Sr concentrations above 150 ppb were observed in 35% of wells, and the Sr/Ca ratio threshold

of 0.006 was exceeded in 72%. Thus, this technique does not provide a conclusive source of Cr(VI) for our samples.

Other techniques relied on threshold values for the Ca/Mg, Ca/Si, Mg/Si, and Na/(Ca+Mg) ratios. In our study, the Ca/Mg ratio was less than 0.33 in 0% of wells, the Mg/Si ratio was greater than 2.2 in only 0.4%, and the Na/(Ca+Mg) ratio was less than 0.33 in only 24.5%. Previous studies attributed Cr(VI) to geogenic sources when Ca/Mg ratios were between 0.01 and 0.33[87, 170, 268] when Na/(Ca+Mg) ratios were less than 0.3,[170] and when Mg/Si ratios were greater than 2.2.[104] Our results are not in keeping with these prior studies, suggesting Cr(VI) was not geogenic in our study. However, the use of unreported water softeners in our study (17.4% of user reported) could have altered Ca or Na concentrations, rendering this method ineffective for treated tap water sampling.

No Cr(VI) samples in this study exceeded the 50 ppb threshold above which contamination is expected to be primarily anthropogenic.[88, 326] This technique was unable to confirm the presence of anthropogenic Cr(VI).

Predictive modeling

When evaluating all data, conductivity ($p<0.05$) and samples from the Piedmont and Blue Ridge aquifer were significant predictors of Cr(VI) (Table H.5). Focusing only on samples from the Piedmont and Blue Ridge, only conductivity was predictive (Table H.6). These results are in agreement with Coyte 2020[79] who found generally higher probabilities of exceedance in the Piedmont and Blue Ridge than other aquifers and no bivariate relationship between Cr(VI) and pH. This lack of relationship was unexpected because pH and DO (not measured in our study) could serve as proxies for redox conditions and water rock interactions. In keeping, both studies also observed a negative relationship between Cr(VI) and

conductivity, which may be attributed to groundwater geochemical parameters.[79] Overall, this technique suggested geogenic contributions from the Piedmont and Blue Ridge aquifer.

G.4 Discussion

The techniques used in our study did not provide a consistent, definitive conclusion on the sources of Cr(VI) in our study area. While geogenic Cr(VI) has previously been identified in the Piedmont and Blue Ridge aquifer,[79, 352] only three techniques identified geogenic sources as a potential source, two techniques indicated that geogenic Cr(VI) was likely not the primary source, and three techniques were inconclusive (Table G.1). Identifying anthropogenic sources was similar, as three suggested anthropogenic sources but five were inconclusive. Specifically looking at coal ash, two techniques did not support coal ash as a primary source and three were inconclusive. The number of inconclusive results could partially be explained by the techniques being originally developed in locations outside North Carolina (e.g., China, Egypt, Nigeria, Greece and Italy) and/or could also be due to the multiple potential Cr(VI) sources in our study location. In addition to evaluating different Cr(VI) sources, we considered three coal ash sources in our study: coal ash impoundments associated with power plants, landfills containing coal ash, and coal ash used as structural fill. While locations of impoundments and landfills are known,[234] documentation of coal ash structural fill locations is incomplete.[227] The NCDEQ maintains a GIS portal of structural fill locations, but they note potential issues with data accuracy and completeness, as coal ash was widely used in the 1980s and 1990s before the implementation of reporting requirements in 1994.[227]

The source tracking techniques we identified primarily focused on regional scale identification. At such a resolution, these approaches require having data from multiple aquifer

systems, potential anthropogenic Cr(VI) sources, and aquifer material. The NCDEQ was able to conclusively confirm coal ash contamination at the Sutton Steam Plant (Wilmington, NC) in 2015 after 6 years of monitoring.[233] They routinely measured As, Tl, TDS, B, Fe, Mn, and Se (Cr(VI) was not included) in monitoring wells immediately upstream and downstream of the coal ash impoundment. The scope, cost, and expertise of this investigation is likely beyond the ability of residents and even local health departments. For well users, such data may be difficult to obtain as many do not know specific characteristics of their wells.[256, 324] Even state or local health departments do not always have such information readily accessible.[210]

Understanding the source(s) of contamination in well water is important, as there are various funding mechanisms available to help users access safe drinking water. In the aftermath of natural disasters, disaster relief funding may be available to well users if their system has been damaged during the event.[124] However, as well users infrequently test their drinking water, especially for emerging contaminants,[50, 256, 324] pre-hurricane baseline data are often not available. Thus, source-tracking techniques can serve to determine contamination source(s) impacting an individual well. Under routine conditions in North Carolina, the Bernard Allen Memorial Emergency Drinking Water Fund (administered by the NCDEQ) provides remediation funding for low-income households with private well contamination by anthropogenic sources.[1] In 2020, reverse osmosis units were installed in 12 homes supplied by private wells when elevated concentrations of As and V were found in Randolph County, and contamination from nearby coal ash was the suspected cause.[229] However, NCDEQ investigations later ruled the contamination was of natural origin by taking water, bedrock, and sediment samples immediately upstream and downstream of the potential contamination source. As a result, device maintenance was no longer covered by the Bernard Allen Fund and some residents opted for NCDEQ to remove their devices.

Overall, our study highlights a critical need to develop and/or refine standardized source tracking techniques that can be used at the household and regional-scale to understand Cr(VI) source(s). While Cr(VI) has become a contaminant of concern in NC, other emerging contaminates (e.g., nanomaterials, plasticizers) also pose risks to drinking water resources, and more acutely, private wells. Like Cr(VI), there is insufficient knowledge on the prevalence, source identification, remediation, and health effects for many emerging contaminants.[127] To ensure well users have access to a safe drinking water supply, data-driven and research-verified information and resources are needed to empower residents to make informed decisions to protect their health.

Table G.1: **Summary of source tracking techniques** applied and whether they were useful in determining source of Cr(VI) in 2018-209 NC private well sampling campaign.

General Approach	Technique	Geogenic	Anthropogenic	
			Agriculture and Industrial	Coal ash disposal site
Geologic Approach	Analyzed published data regarding rock types in region	Potential sources, wells in Piedmont and Blue Ridge aquifer and with known mafic rocks had higher Cr(VI) concentrations	-	-
Spatial Analysis	Mapped Cr(VI) values relative to potential sources	-	-	Not primary source, lower Cr(VI) concentrations around sites
	Evaluated distance between measured Cr(VI) value and potential sources	-	-	Not primary source Cr(VI) not correlated with distance to site
	Mapped Cr(VI) values relative to published land-use data in the region	Not primary source, Cr(VI) lower in undeveloped land	Potential source, higher Cr(VI) in developed and agricultural land	-
Correlations	Correlations between all measured parameters	Inconclusive, expected correlations not observed		
	Correlations between Cr(VI) and V, SO_4^{2-}, Cl, and Si	Potential source, moderate to strong correlations observed	-	-
	Correlations between Cr(VI) and Co, Fe, Mn, Na, and Se	Inconclusive expected correlations not observed	-	-
	Correlations between Cr(VI) and NO_3^-	-	Potential source, weak correlation observed	-
	Correlations between Cr(VI) and P, B	-	Inconclusive, expected correlations not observed	-
	Correlations between Cr(VI) and Mg, Ni, and B	-	Inconclusive, expected correlations not observed	-
Statistical Analysis	Performed principal component analysis of water quality parameters	Inconclusive, no previously identified groupings observed		

Table G.1: **Summary of source tracking techniques** applied and whether they were useful in determining source of Cr(VI) in 2018-209 NC private well sampling campaign.

General Approach	Technique	Geogenic	Anthropogenic	
			Agriculture and Industrial	Coal ash disposal site
	Performed cluster analysis of water quality parameters	-	Potential source, groupings of Cr(VI), NO_3^-, and P observed	-
Thresholds	Established coal ash thresholds for B and Sr, and B/Cr and Sr/Ca ratios	-	-	Inconclusive, not all thresholds exceeded
	Compared Ca/Mg, Ca/Si Mg/Si, and Na/(Ca+Mg) ratios	Not primary source, not all thresholds exceeded	-	-
	Used 100-150 ppb as rough geogenic Cr(VI) threshold	-	Inconclusive, no samples > 50 ppb	-
Modeling	Tobit regression model of field parameters and aquifer type	Potential source, Piedmont and Blue Ridge aquifer was predictive of Cr(VI)	-	-

G.5 Acknowledgements

The research was supported by the National Science Foundation through a Rapid Research Response grant (#1855567), Spring Point Partners, and the UNC Superfund Research Program (P42-ES031007). We would like to thank our community partners, which includes the Blue Ridge Environmental Defense, Cape Fear and Lumbee Riverkeepers, Chatham and Northampton County Citizens Against Coal Ash, Lumbee Tribe Administration, Lumbee Tribe Veterans Resource Center, Iredell County Emergency Management Division, Iredell County Health Department, New Hanover County and Robeson County Cooperative Extension, Rocky Mount Church in Mooresville, and Winyah Rivers Alliance. We would also like to thank support of faculty and students at Virginia Tech and the University of North Carolina at Chapel Hill's Institute for the Environment for their assistance in sampling. Lastly,

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Appendix H

Supplemental Information for Appendix G

H.1 Supplemental Tables for Appendix G

Table H.1: **Description of general approaches and specific techniques used in literature for the source identification of Cr(VI) in groundwater study**

General Approach (# papers using approach)	Description	# Unique techniques (# used in this study)	Techniques*	# papers using technique
Geologic setting (45)	Evaluation of aquifer materials and their potential release to groundwater	3 (1)	**Analyzed published data regarding rock types in region**	40 [42, 43, 54, 67, 79, 85, 86, 87, 104, 122, 138, 140, 143, 165, 168, 170, 174, 175, 189, 211, 217, 222, 223, 245, 246, 247, 250, 268, 277, 278, 287, 326, 331, 340, 350, 352, 357, 369]
			Analyzed Cr content of samples of aquifer material	19 [42, 52, 87, 88, 104, 122, 136, 140, 150, 168, 174, 175, 201, 217, 222, 245, 277, 278, 354]
			Performed leaching tests to determine potential of aquifer materials to release Cr(VI)	8 [43, 87, 104, 175, 201, 222, 278, 354]
Spatial analysis (41)	Analysis of location and distance of Cr(VI) concentrations relative to potential sources	3 (3)	**Mapped Cr(VI) values relative to potential sources**	40 [54, 67, 79, 86, 88, 136, 138, 143, 150, 151, 161, 165, 168, 170, 174, 175, 189, 194, 201, 211, 217, 222, 245, 246, 247, 250, 277, 287, 288, 311, 317, 326, 331, 340, 349, 350, 352, 354, 357, 369]
			Evaluated distance between measured Cr(VI) value and potential sources	6 [43, 88, 143, 168, 201, 352]
			Mapped Cr(VI) values relative to published land-use data in the region	5 [165, 217, 311, 331, 340]
Correlations (30)	Identification of correlations between Cr(VI) and other water quality parameters	10 (8)	*These studies performed correlations between:*	
			Cr(VI) and total Cr	15 [79, 86, 88, 104, 122, 136, 165, 170, 175, 245, 250, 340, 349, 350, 369]
			All measured parameters including Cr(VI)	8 [161, 175, 194, 223, 246, 331, 349, 369]
			Identifying natural occurrence:	
			Cr(VI) and Si, Ca, Ni, Fe, Mg, HCO_3^-	10 [43, 86, 143, 165, 175, 223, 245, 246, 250, 350]

Table H.1: **Description of general approaches and specific techniques used in literature for the source identification of Cr(VI) in groundwater study**

General Approach (# papers using approach)	Description	# Unique techniques (# used in this study)	Techniques*	# papers using technique
			Cr(VI) and Mn to determine natural oxidation of Cr(III) by Mn oxides	5 [136, 140, 174, 175, 288]
			Cr(VI) and V and Se to determine similar natural oxidation/reduction pathways	2 [86, 165]
			Cr(VI) and Na^+ and Ca^{2+} to determine natural ion exchange in aquifers resulting in Cr(VI) desorption	4 [52, 174, 194, 357]
			Identifying anthropogenic contamination:	
			Cr(VI) and NO_3^-, NH_3, DDE or P to determine agricultural sources	12 [54, 67, 143, 150, 165, 189, 211, 217, 247, 288, 317, 350]
			Cr(VI) and chlorinated solvents (i.e. DCE) to determine industrial contamination	1 [143]
			Cr(VI) Mg, Ni, and B within plume	1 [86]
			Cr(VI) and Cl^-, SO_4^{2-}, HCO_3^-, Ca^{2+} to determine natural Cr(VI) as opposed to anthropogenic ions	5 [67, 168, 189, 194, 357]
Multivariate analysis (13)	Multivariate statistical analysis of common water quality parameters including Cr(VI)	3 (2)	**Performed principal component analysis of water quality parameters**	9 [165, 217, 222, 246, 287, 326, 331, 350, 369]
			Performed factor analysis using physical and chemical parameters of water samples	7 [175, 217, 246, 247, 317, 326, 350]

Table H.1: **Description of general approaches and specific techniques used in literature for the source identification of Cr(VI) in groundwater study**

General Approach (# papers using approach)	Description	# Unique techniques (# used in this study)	Techniques*	# papers using technique
			Performed cluster analysis of water quality parameters	5 [143, 246, 247, 350, 369]
Thresholds (33)	Comparison of water quality parameters with specific threshold values	7 (3)	**Established coal ash contamination thresholds for B and Sr, and B/Cr and Sr/Ca ratios**	1 [352]
			Compared pH, DO, water quality, and redox potential from different wells to establish relative water age	12 [86, 138, 165, 170, 211, 268, 277, 288, 326, 331, 340, 354]
			Compared Ca^{2+}/ HCO_3^- ratios between wells to determine relative depth of groundwater	1 [86]
			Compared ternary diagrams of major ions of differing groundwater types	22 [52, 54, 87, 122, 150, 168, 170, 189, 194, 211, 246, 247, 250, 268, 278, 288, 317, 331, 349, 350, 354, 357]
			Compared HCO_3^-/Mg, Ca/Mg, SiO_2/HCO_3^-Ca/Si, and Mg/Si ratios with those observed in groundwater containing known geogenic Cr(VI)	8 [86, 104, 170, 189, 223, 247, 268]
			Compared Cr(VI) results to rough geogenic thresholds of 50-100 ppb	2 [88, 326]
			Compared distribution of observed data to a normal distribution	2 [67, 340]
Isotope ratio (6)	Use of isotopic ratios to gain insight into potential Cr(VI) or water sources	2 (0)	Determined $^{53}Cr/^{52}Cr$ ratio to examine the degree of reduction of Cr(VI)	5 [104, 151, 168, 175, 354]

Table H.1: **Description of general approaches and specific techniques used in literature for the source identification of Cr(VI) in groundwater study**

General Approach (# papers using approach)	Description	# Unique techniques (# used in this study)	Techniques*	# papers using technique
			Compared $^{87}Sr/^{86}Sr$ ratios to differentiate natural and anthropogenic contamination	1 [352]
Predictive modeling (17)	Use of modeling software to understand and predict contaminant flow and determine if hypothesized sources are valid	5 (1)	Modeled aquifer materials and solute conditions to estimate potential for geogenic Cr(VI) release	5 [42, 87, 122, 189, 222]
			Modeled chemical reactions occurring in drinking water to estimate solubility and speciation of Cr(VI)	8 [52, 54, 104, 138, 168, 317, 331, 354]
			Constructed regression model from common field parameters (pH, TDS, DO) to estimate probability of high geogenic Cr(VI)	1 [79]
			Developed geostatistical modeling to identify clusters of high concentrations of water quality parameters	3 [168, 311, 350]
			Conducted groundwater modeling to simulate Cr(VI) variability due to observed results	2 [174, 222]

*Bold text indicates techniques used in this study

Table H.2: **Summary of Cr(VI) sources identified in the literature**

Cr(VI) source	General approaches used
All (n=51)	Geologic setting (n=45) [42, 43, 52, 54, 67, 79, 86, 87, 88, 104, 122, 136, 138, 140, 143, 150, 165, 168, 170, 174, 175, 189, 201, 211, 217, 222, 223, 245, 246, 247, 250, 268, 277, 278, 287, 288, 317, 326, 331, 340, 350, 352, 354, 357, 369] Spatial analysis (n=41) [43, 54, 67, 79, 87, 88, 136, 138, 143, 150, 151, 161, 165, 168, 170, 174, 175, 189, 194, 201, 211, 217, 222, 245, 246, 247, 250, 277, 287, 288, 311, 317, 326, 331, 340, 349, 350, 352, 354, 357, 369] Thresholds (n=33) [52, 54, 67, 86, 87, 88, 104, 122, 138, 150, 165, 168, 170, 189, 194, 211, 223, 246, 247, 250, 268, 277, 278, 288, 317, 326, 331, 340, 349, 350, 352, 354, 357] Correlations (n=30) [43, 52, 54, 67, 86, 136, 140, 143, 150, 161, 165, 168, 174, 175, 189, 194, 211, 217, 223, 245, 246, 247, 250, 288, 317, 331, 349, 350, 357, 369] Predictive modeling (n=17) [42, 52, 54, 79, 87, 87, 104, 122, 138, 168, 174, 189, 222, 311, 317, 331, 350, 354] Multivariate analysis (n=13) [143, 164, 175, 217, 222, 246, 247, 287, 317, 326, 331, 350, 369] Isotope ratio (n=6) [104, 151, 168, 175, 352, 354]
Agricultural activity (n=16) [67, 87, 143, 150, 165, 189, 211, 217, 246, 247, 288, 317, 326, 350, 357, 369]	Geologic setting (n=16) [67, 87, 143, 150, 165, 189, 211, 217, 246, 247, 288, 317, 326, 350, 357, 369] Spatial analysis (n=15) [67, 143, 150, 165, 189, 211, 217, 246, 247, 288, 317, 326, 350, 357, 369] Correlations (n=14) [67, 143, 150, 165, 189, 211, 217, 246, 247, 288, 317, 350, 357, 369] Thresholds (n=13) [67, 87, 150, 165, 189, 211, 246, 247, 288, 317, 326, 350, 357] Multivariate analysis (n=9) [143, 165, 217, 246, 247, 317, 326, 350, 369] Predictive modeling (n=4) [87, 189, 317, 350] Isotope ratio (n=0
Geogenic (n=44) [42, 43, 52, 54, 67, 79, 86, 87, 88, 104, 122, 136, 138, 140, 143, 150, 165, 168, 170, 174, 175, 189, 194, 211, 217, 222, 223, 245, 246, 247, 250, 268, 277, 278, 287, 288, 311, 317, 326, 340, 350, 352, 357, 369]	Geologic setting (n=42) [42, 43, 52, 54, 67, 79, 86, 87, 88, 104, 122, 136, 138, 140, 143, 150, 165, 168, 170, 174, 175, 189, 211, 217, 222, 223, 245, 246, 247, 250, 268, 277, 278, 287, 288, 317, 326, 340, 350, 352, 357, 369] Spatial analysis (n=35) [43, 54, 67, 79, 86, 88, 104, 122, 136, 138, 143, 150, 165, 168, 170, 174, 175, 189, 211, 217, 222, 245, 246, 247, 250, 277, 287, 288, 311, 317, 326, 340, 350, 352, 357, 369] Thresholds (n=30) [52, 54, 67, 86, 87, 88, 104, 122, 138, 150, 165, 168, 170, 189, 194, 211, 223, 246, 247, 250, 268, 277, 278, 288, 317, 326, 340, 350, 352, 357] Correlations (n=27) [43, 52, 54, 67, 86, 136, 140, 143, 150, 165, 168, 174, 175, 189, 194, 211, 217, 223, 245, 246, 247, 250, 250, 288, 317, 350, 357, 369] Predictive modeling (n=15) [42, 52, 54, 79, 87, 104, 122, 138, 168, 174, 189, 222, 311, 317, 350] Multivariate analysis (n=12) [143, 165, 175, 217, 222, 246, 247, 287, 317, 326, 350, 369]

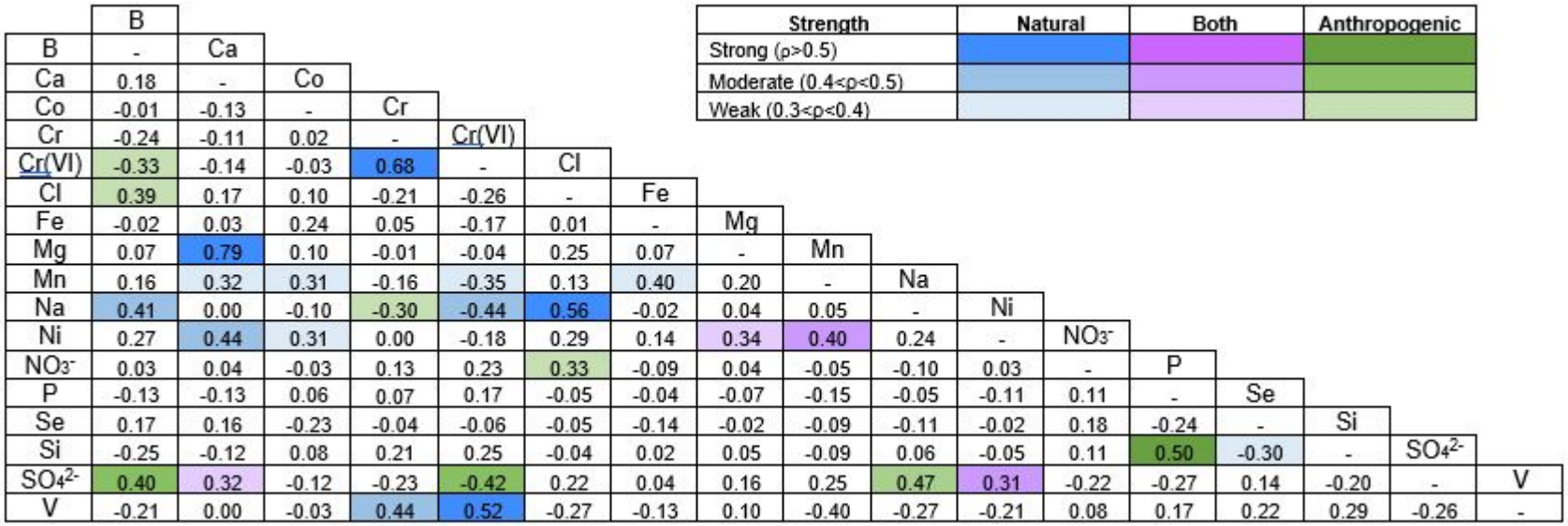
Table H.2: **Summary of Cr(VI) sources identified in the literature**

Cr(VI) source	General approaches used
	Isotope ratio (n=4) [104, 168, 175, 352]
Industrial: coal ash (n=6) [170, 175, 287, 350, 352, 357]	Geologic setting (n=6) [170, 175, 287, 350, 352, 357] Spatial analysis (n=6) [170, 175, 287, 350, 352, 357] Thresholds (n=4) [170, 350, 352, 357] Correlations (n=3) [175, 350, 357] Multivariate analysis (n=3) [175, 287, 350] Isotope ratio (n=2) [175, 352] Predictive modeling (n=1) [350]
Industrial: non-coal ash (n=28) [43, 86, 88, 104, 136, 143, 151, 161, 162, 168, 194, 201, 217, 222, 223, 245, 287, 288, 311, 317, 326, 331, 340, 349, 350, 354, 357, 369]	Spatial analysis (n=25) [43, 86, 88, 136, 143, 151, 161, 168, 194, 201, 217, 222, 245, 287, 288, 311, 317, 326, 331, 340, 349, 350, 354, 357, 369] Geologic setting (n=22) [43, 86, 88, 136, 143, 168, 201, 217, 222, 223, 245, 287, 288, 317, 326, 331, 340, 350, 354, 357, 369] Correlations (n=17) [43, 86, 136, 143, 161, 168, 194, 217, 223, 245, 288, 317, 331, 349, 350, 357, 369] Thresholds (n=15) [86, 88, 104, 168, 194, 223, 288, 317, 326, 331, 340, 349, 350, 354, 357] Multivariate analysis (n=9) [143, 217, 222, 287, 317, 326, 331, 350, 369] Predictive modeling (n=8) [104, 168, 222, 311, 317, 331, 350, 354] Isotope ratio (n=4) [104, 151, 168, 354]

Parameter		Recommended levels			Percent exceeding standard (US EPA/NC 2L)[c]				
		USEPA SDWA	DHHS HRE	DEQ 2L	All aquifers[b]	Castle Hayne	Northern Atlantic	Piedmont & Blue Ridge	Surficial
Exceeded at least one health-based recommended level					5.6%	3.1%	5.3%	5.8%	6.7%
Exceeded at least one aesthetic-based level					46.4%	39.1%	84.1%	40.4%	73.3%
[b]Antimony	Health-based	6 ppb	-	1 ppb	0.0/0.0.08%	0.0/0.0%	0.0/0.0%	0.0/0.1%	0.0/0.0%
[b]Arsenic	Health-based	10 ppb			0.08%	1.6%	0.0%	0.0%	0.0%
[b]Barium	Health-based	2,000 ppb		700 ppb	0.0/0.08%	0.0/0.0%	0.0/0.0%	0.0/0.1%	0.0/0.0%
[b]Boron	Health-based	-	-	700 ppb	0.08%	0.0%	0.0%	0.1%	0.0%
[a]Cadmium	Health-based	5 ppb		2 ppb	0.4/1.0%	NA	0.0/0.0%	0.4/1.0%	NA
[b]Chromium	Health-based	100 ppb		10 ppb	0.0/1.2%	0.0/0.0%	0.0/0.0%	0.0/1.5%	0.0/0.0%
[b]Cr(VI)*	Health-based	-	0.07 ppb	-	62.0%	21.8%	20.5%	72.0%	16.7%
[b]Cobalt	Health-based	-	-	1 ppb	5.6%	4.7%	38.4%	0.6%	13.3%
[a]Copper	Health-based	1,300 ppb		1,000 ppb	12.3/15.7%	NA	16.7/17.8%	11.8/15.4%	NA
[b]Fluoride	Health-based	4 ppm		2 ppm	0.08/1.3%	0.0/1.6%	0.7/0.7%	0.0/1.5%	0.0/0.0%
[a]Lead	Health-based	15 ppb			8.5%	NA	13.3%	8.0%	NA
[a]Nickel	Health-based	-	-	100 ppb	1.7%	NA	3.3%	1.5%	NA
[b]Selenium	Health-based	50 ppb		20 ppb	0.0/0.0%	0.0/0.0%	0.0/0.0%	0.0/0.0%	0.0/0.0%
[b]Thallium	Health-based	-	-	2 ppb	0.0%	0.0%	0.0%	0.0%	0.0%
[b]Uranium	Health-based	30 ppb	-	-	1.1%	0.0%	0.0%	1.4%	0.0%
[b]Vanadium	Health-based	-	-	7 ppb	11.7%	1.6%	0.7%	14.4%	0.0%
[b]Chloride	Aesthetic-based	250 ppm			0.2%	1.6%	0.0%	0.2%	0.0%
[b]Iron	Aesthetic-based	300 ppb			10.2%	21.9%	27.2%	6.4%	30.0%
[b]Manganese	Aesthetic-based	50 ppb			7.9%	10.9%	17.9%	5.8%	23.3%
[b]pH	Aesthetic-based	6.5-8.5	-	6.5-8.5	36.0%	14.0%	73.5%	31.2%	56.7%
[b]Sulfate	Aesthetic-based	250 ppm			1.0%	0.0%	0.0%	1.2%	0.0%
[b]TDS	Aesthetic-based	500 ppm	-	500 ppm	3.1%	10.9%	2.0%	2.7%	6.7%
[a]Zinc	Aesthetic-based	5 ppm		1 ppm	4.7/27.8%	NA	4.4/27.8%	4.6/27.8%	NA

USEPA: U.S. Environmental Protection Agency
SDWA: Safe Drinking Water Act
DHHS: Department of Health and Human Services
NCDEQ: NC Department of Environmental Quality
Cr(VI): Hexavalent chromium
TDS: Total dissolved solids
*Hexavalent chromium has a public health goal established by NC Department of Health and Human Services
[a]Total concentration measured in the first draw
[b]Total concentration measured in the flushed sample
[c]Sample size:
First draws: All, n=1,110; Castle Hayne, n=0; Northern Atlantic, n=90; Piedmont and Blue Ridge, n=1,015; Surficial Aquifer, n=0
Flushed: All, n=1,265; Castle Hayne, n=60; Northern Atlantic, n=149; Piedmont and Blue Ridge, n=1,015; Surficial Aquifer, n=28

Figure H.1: **Summary of water quality results for the 2018-2019 NC private well water sampling campaign**



	B																
B	-	Ca															
Ca	0.18	-	Co														
Co	-0.01	-0.13	-	Cr													
Cr	-0.24	-0.11	0.02	-	Cr(VI)												
Cr(VI)	-0.33	-0.14	-0.03	0.68	-	Cl											
Cl	0.39	0.17	0.10	-0.21	-0.26	-	Fe										
Fe	-0.02	0.03	0.24	0.05	-0.17	0.01	-	Mg									
Mg	0.07	0.79	0.10	-0.01	-0.04	0.25	0.07	-	Mn								
Mn	0.16	0.32	0.31	-0.16	-0.35	0.13	0.40	0.20	-	Na							
Na	0.41	0.00	-0.10	-0.30	-0.44	0.56	-0.02	0.04	0.05	-	Ni						
Ni	0.27	0.44	0.31	0.00	-0.18	0.29	0.14	0.34	0.40	0.24	-	NO_3^-					
NO_3^-	0.03	0.04	-0.03	0.13	0.23	0.33	-0.09	0.04	-0.05	-0.10	0.03	-	P				
P	-0.13	-0.13	0.06	0.07	0.17	-0.05	-0.04	-0.07	-0.15	-0.05	-0.11	0.11	-	Se			
Se	0.17	0.16	-0.23	-0.04	-0.06	-0.05	-0.14	-0.02	-0.09	-0.11	-0.02	0.18	-0.24	-	Si		
Si	-0.25	-0.12	0.08	0.21	0.25	-0.04	0.02	0.05	-0.09	0.06	-0.05	0.11	0.50	-0.30	-	SO_4^{2-}	
SO_4^{2-}	0.40	0.32	-0.12	-0.23	-0.42	0.22	0.04	0.16	0.25	0.47	0.31	-0.22	-0.27	0.14	-0.20	-	V
V	-0.21	0.00	-0.03	0.44	0.52	-0.27	-0.13	0.10	-0.40	-0.27	-0.21	0.08	0.17	0.22	0.29	-0.26	-

Strength	Natural	Both	Anthropogenic
Strong (ρ>0.5)			
Moderate (0.4<ρ<0.5)			
Weak (0.3<ρ<0.4)			

Figure H.2: **Correlation matrix of all parameters in Section G.3.2.**
Correlations are grouped based on potential sources identified in the literature and correlations strength

Table H.3: **Results of k-means clustering showing the differences in median values of parameters between assigned groups**

Parameter	Group 1	Group 2	Group 3	Group 4
Number of wells	62	504	21	145
Cr(VI) (ppb)	0.06	0.55	0.07	0.08
NO_3^- (ppm)	0.12	0.88	0.24	0.12
P (ppb)	13.3	41.1	33.1	5.6

Table H.4: **Comparison of water values from 2018-2019 NC private well sampling campaign to coal ash threshold parameters discussed in literature**

Parameters or ratio	Thresholds	Wells exceeding threshold in Piedmont and Blue Ridge[a]		Interpretation of threshold
		# wells	% wells	
B	>100 ppb	2	0.20%	Samples are impacted by coal ash
B/Cr	>200	7	0.69%	
Sr	>150 ppb	354	34.88%	
Sr/Ca	0.006	734	72.32%	
Ca/Mg	>1.0	957	94.29%	Lack of interaction with mafic rocks
Ca/Mg	>1.0	0	0.00%	Interactions with mafic rocks
Mg/Si	>2.2	4	0.39%	
Na/(Ca+Mg)	<0.33	249	24.53%	

[a]n=1015

Table H.5: **Results of Tobit regression model predicting Cr(VI) concentrations based on conductivity and pH by aquifer**

Parameter	Estimate	Std. error	t value	Pr(>t)	
Intercept	-1.64E-01	3.77E-01	-0.435	0.6632	
Conductivity	-4.65E-04	9.82E-05	-4.736	2.18E-06	***
pH	1.25E-05	2.05E-05	0.61	0.5418	
Castle Hayne	-3.33E-01	4.23E-01	-0.786	0.4317	
Northern Atlantic	-3.9eE-01	4.06E-01	-0.965	0.3345	
Piedmont and Blue Ridge	8.47E-01	3.77E-01	2.245	0.0248	*
Surficial	-5.73E-01	4.73E-01	-1.213	0.2251	
logSigma	2.08E-01	2.26E-02	9.218	<2E-16	***

Table H.6: **Results of Tobit regression model predicting Cr(VI) concentrations based on conductivity and pH in the Piedmont and Blue Ridge aquifer**

Parameter	Estimate	Std. error	t value	Pr(>t)	
Intercept	9.35E-01	6.22E-02	15.304	< 2E-16	***
Conductivity	-1.55E-03	2.03E-04	-7.605	2.85E-14	***
pH	3.98E-05	4.00E-05	0.994	0.32	
logSigma	2.34E-01	2.38E-02	9.848	<2E-16	***

H.2 Supplemental Figures for Appendix G

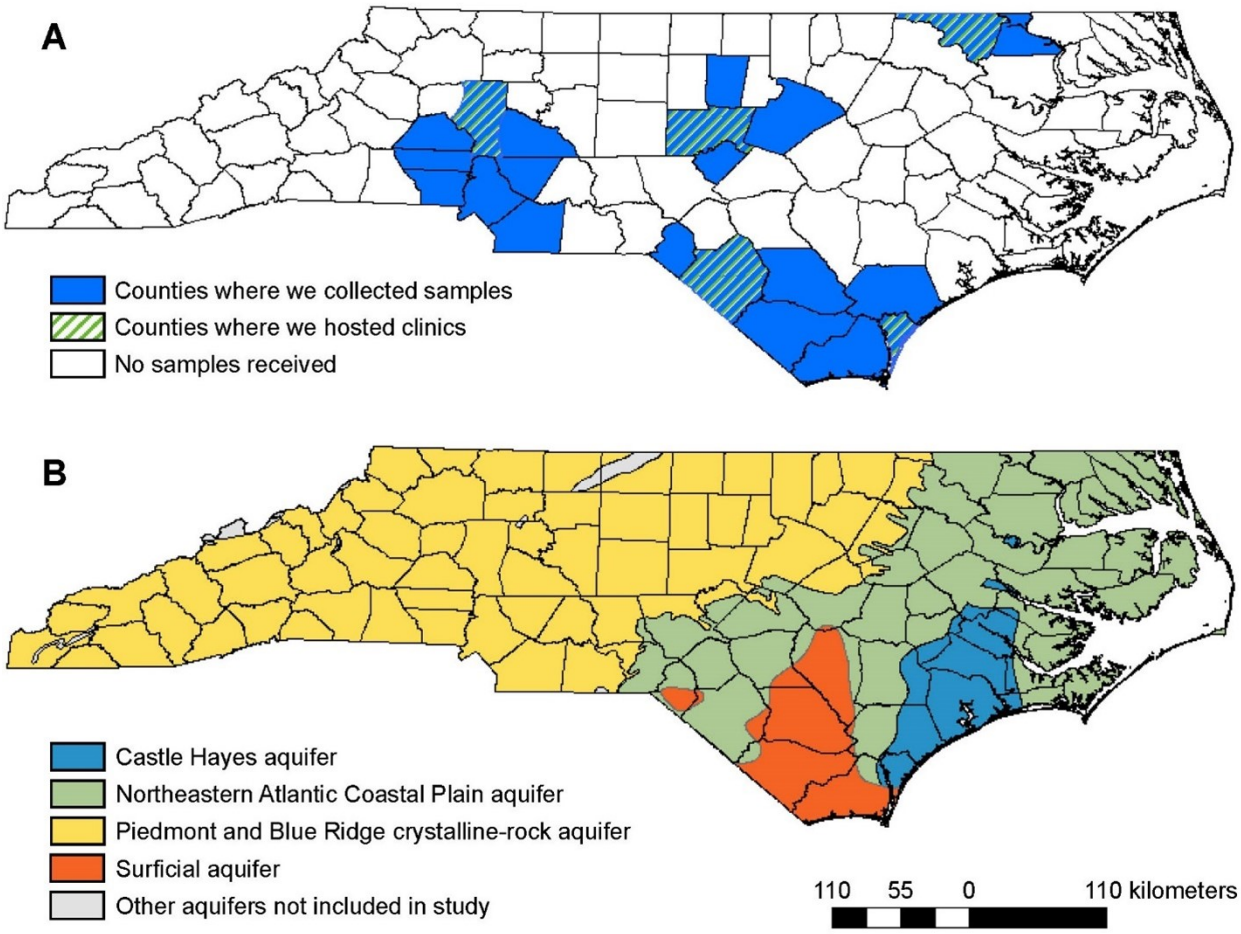


Figure H.3: **NC Citizen Science Sampling**
(A) Counties where we hosted and samples were collected and (B) the primary aquifers as designated in the USGS Ground Water Atlas[6]

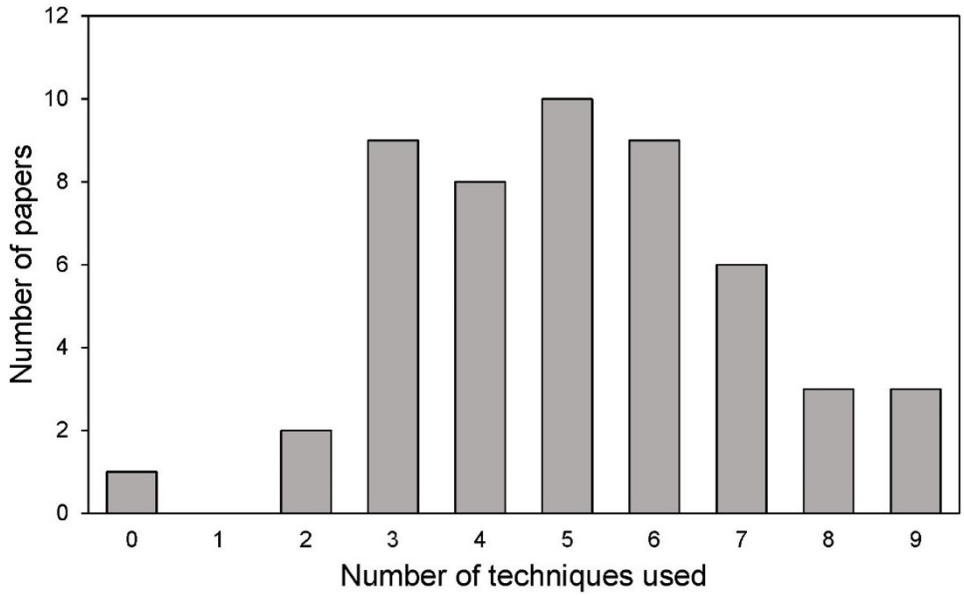


Figure H.4: **Number of Cr(VI) source tracking techniques used per paper evaluated.**

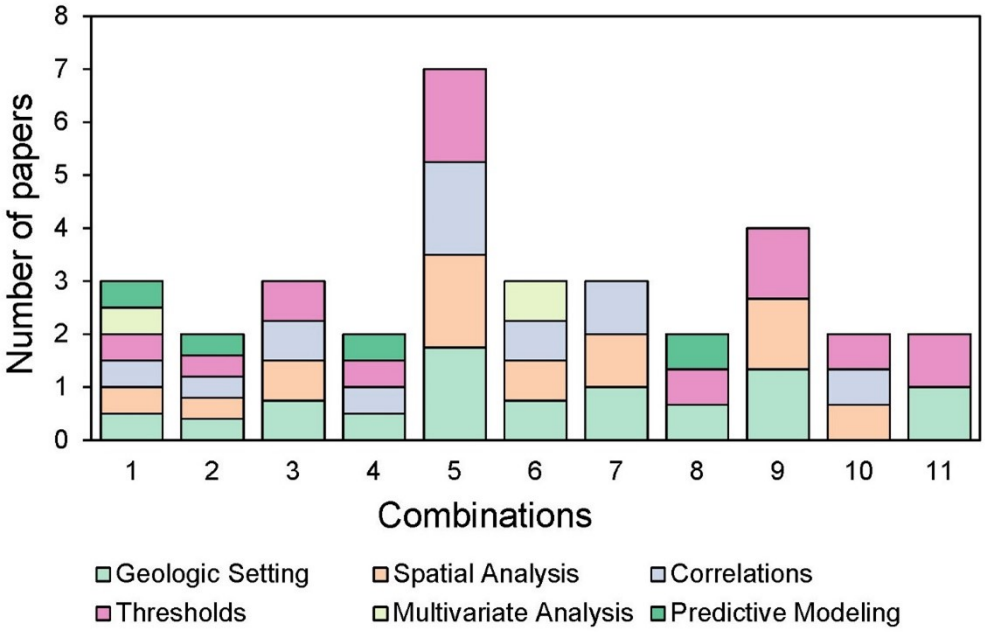


Figure H.5: **Combinations of general approaches and number of publications using the combination.**
Note: 18 additional combinations of approaches were used but not shown as they were only used in one paper.



Figure H.6: **Kriging interpolation of Cr(VI) concentrations** from private wells sampled in the Piedmont and Blue Ridge aquifer based on distance to the (A) Marshall Steam Station in Iredell and the (B) Brickhaven Mine and Cape Fear Plant in Chatham.

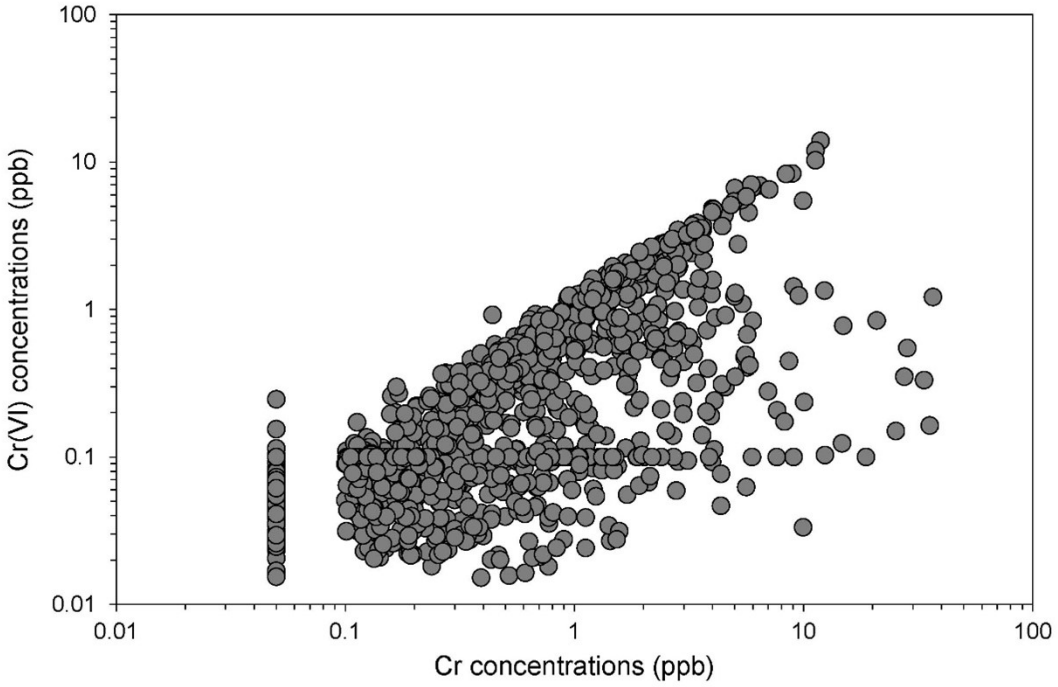


Figure H.7: **Paired analysis of total Cr concentrations compared to Cr(VI) concentrations**

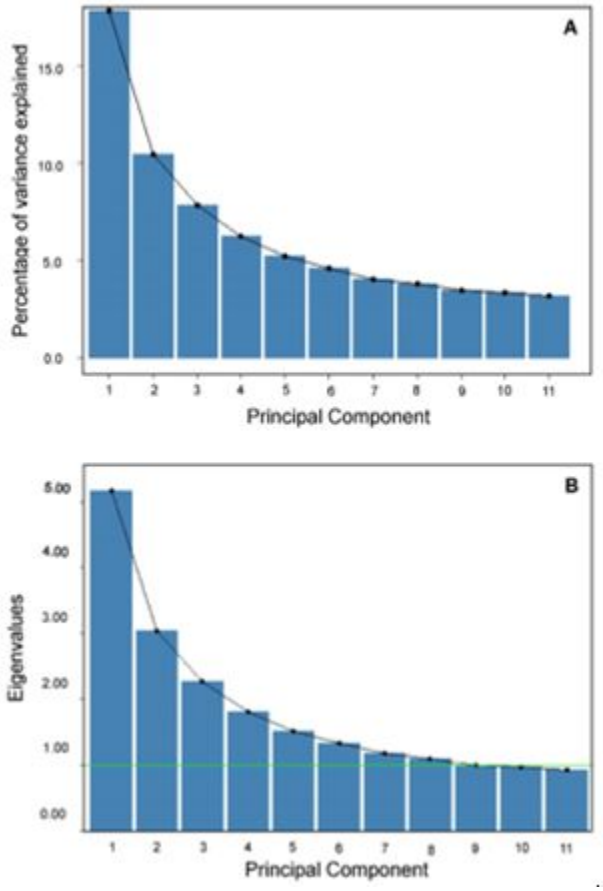


Figure H.8: **Scree plots from PCA analysis** showing (A) percentage of variance explained by each component and (B) the number of principal components with eigenvalues greater than 1.0 (green line)

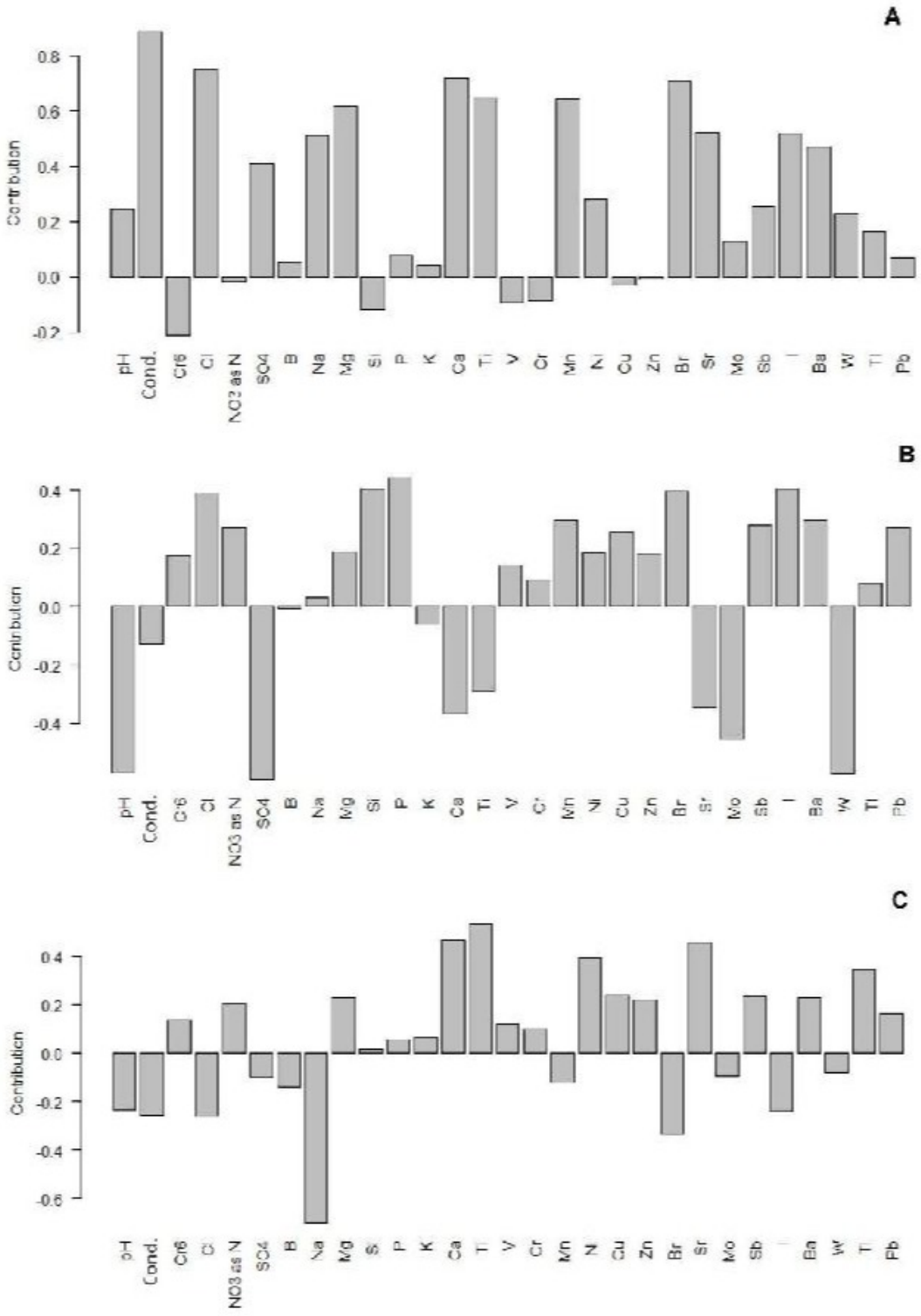


Figure H.9: **Summary of PCA results showing the variable loadings for** (A) first principal component axis, (B) second principal component axis, and (C) third principal component axis.

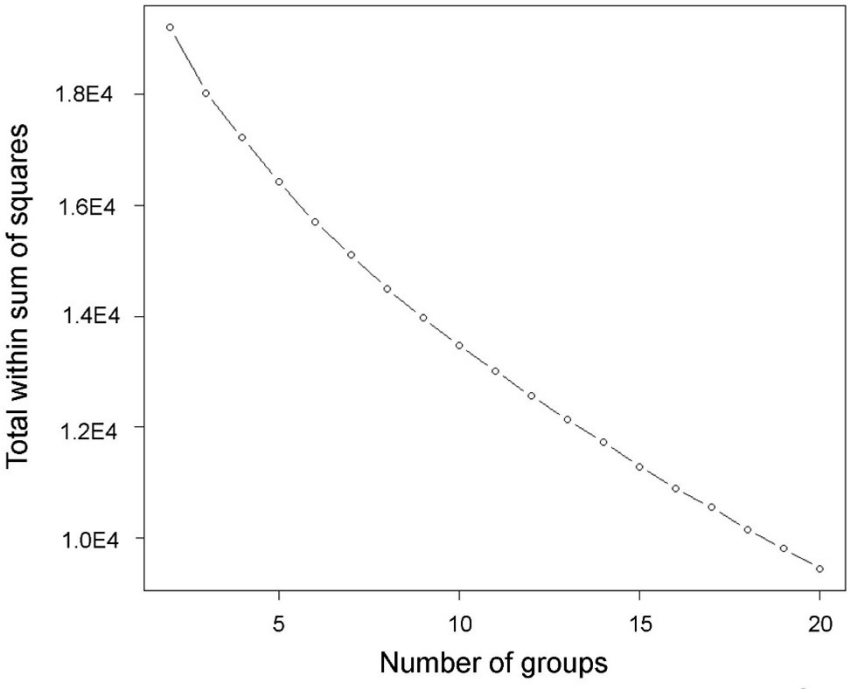


Figure H.10: **Sum of the sum of squares residuals within each group for various number of groups in K-means cluster analysis.**

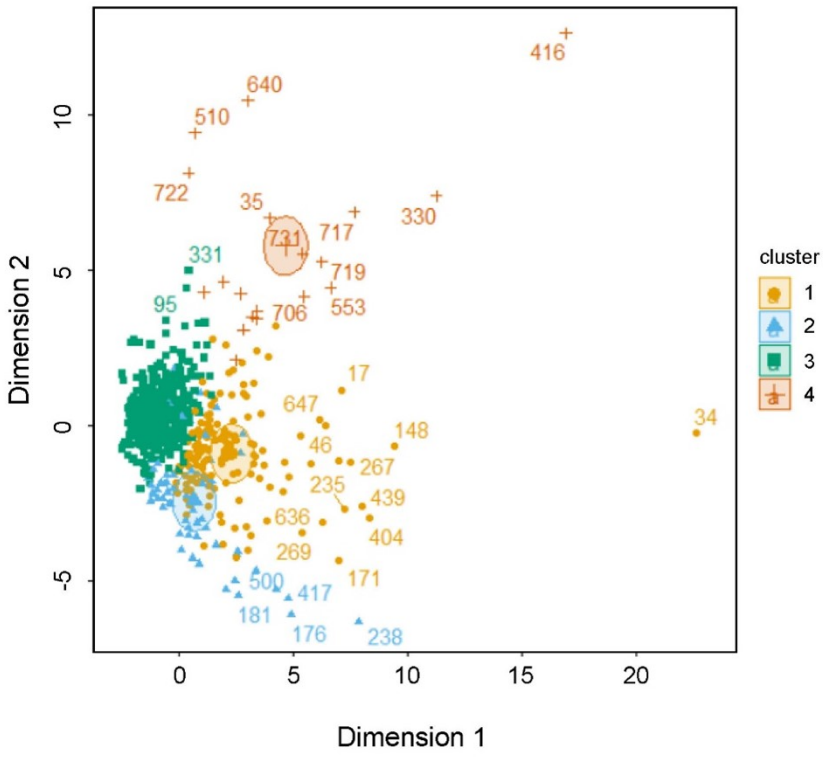


Figure H.11: **Grouping derived from k-means clustering results.**

Appendix I

Experiences in Citizen Science Related to Consumer Perceptions of Water Safety (2011-2022)

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Abstract

Citizen science is utilized by the water industry to collect samples under the provisions of the U.S. EPA Lead and Copper Rule (LCR). Interestingly, informed citizen scientists in Washington D.C. who sampled water for lead, became distrustful of official pronouncements of safe drinking water and eventually helped expose the 2004 Washington D.C. lead crisis. That experience set the stage for use of citizen science in Flint, MI and St. Joe, LA to contradict official messaging regarding water safety, which led to declared water emergencies and financial relief. Our research team has continued engaging in citizen science projects in various cities and herein we document: 1) our process for engaging citizens, 2) a summary of outcomes for a small percentage of cases that found noteworthy problems, and 3) lessons learned.

I.1 Introduction

The promise of citizen science is of growing interest.[53, 330] Its practitioners have a diversity of approaches and goals,[94, 230] which have prompted discussions of best practices and appropriate rules of engagement.[56, 94, 235, 338] At one extreme, citizen science is the only approach that can be used by scientists to study certain problems.[46, 74] Another extreme takes on projects to address concerns of social justice,[46] such as sampling to provide a check on the normal authorities in addressing environmental problems or address power differentials.[242, 281, 310] Sometimes a project designed to address consumer concerns evolves into a project with social justice concerns, as in the case of our team's work helping to expose the Flint Water Crisis in 2016.

Citizen science for regulatory purposes was first utilized by the water industry in 1991 to gather samples for the Lead and Copper Rule (LCR), which requires first-draw regulatory compliance samples to be collected from single family homes with high-risk lead plumbing by utility personnel or consumers.[75] Most utilities opt to have consumers collect these samples using bottles and instructions provided by the water utility.

Weaknesses in this approach became obvious in 2000 after a well-intended switch from chlorine to chloramine combined with a failure to implement optimized corrosion control, triggered the worst lead drinking water contamination event in modern U.S. history in Washington D.C. The problem was not widely publicized until a front page Washington Post expose in January 2004.[109, 117] Two employees of the water utility coordinating the collection of samples with residents were fired in 2001 and 2003 after reports of sampling misconduct[7]. That eventually led to a successful whistleblower lawsuit by one fired employee[355]. The water utility continued to use questionable sampling practices that could have hidden ongoing problems through at least 2008, which resulted in a fine of the utility by the EPA in 2007,[308] and prompted pleas for independent citywide sampling to verify the utilities claims of compliance that never came to pass[10, 117]. The scale and magnitude of the health harm from the D.C. Lead Crisis were not proven until 2009-2013.[55, 107, 192]

All of these experiences set the stage for a water lead crisis in Flint, MI from 2014-2016, that was exposed by a collaboration between our team and citizens. That work ultimately resulted in a declared federal emergency and over one billion dollars in state and federal funding along with legal settlements to the financially beleaguered city.[2, 323]

Our research team at Virginia Tech with other collaborators at Louisiana State University thereafter used citizen science to expose the citywide problems with water lead in St. Joseph, LA and elsewhere. Here, we summarize our experiences addressing citizen concerns related to their potable water from 2011-present, resulting from about 100 resident complaints about

water quality in which we coordinated water sampling events across the United States. Most engagements were instigated by consumers who were worried about the safety of their drinking water, and who for whatever reason, did not completely trust the reassurances of the relevant scientific authorities. This paper is aimed at documenting: 1) our process for engaging citizens, 2) a summary of outcomes relevant to consumers, and 3) some lessons learned.

I.2 Methods

Cases were identified in which citizens concerned about varying aspects of their water quality contacted co-authors Edwards and Katner. While many citizens expressed concerns about more than one contaminant, regulatory issue or perceived personal health ailment, the case studies discussed herein were only those for which samples were collected and analyzed. Information about the sampling intervention, including the tested analytes, how participants were chosen and/or recruited was summarized for select cases (Table I.1). Approximate costs were estimated for each intervention using discretionary research funds donated to the university for that purpose. The degree to which citizens could collect specialized samples versus trained scientists varied as did outcomes. Analytical methods have been reported elsewhere[259, 285, 306].

A template was developed to succinctly present the background information, analytes sampled, and outcomes. Background information can include a summary of the citizen concerns, personal experiences, laboratory data, or actions they had taken to try to resolve the concerns with relevant authorities. Information about the sampling intervention included the tested analytes, how participants were chosen and/or recruited, and the degree to which citizens could collect samples versus trained scientists from the university. The results of each

sampling campaign were documented, including how they were communicated to residents, and with the community, media, or regulatory authorities. Outcomes were highly variable.

I.3 Case Studies

I.3.1 Flint, MI

Background: In 2011, the city of Flint (pop. 96,000) fell under state control after going bankrupt. For a variety of reasons, including a desire to save money, state emergency managers and others decided to switch from purchasing treated Lake Huron water from Detroit to producing their own water using a local treatment plant drawing from the nearby Flint River in April 2014. Federally mandated corrosion control measures were not implemented. The change in source water and lack of corrosion control resulted in on-going customer complaints about the smell, taste, and appearance of the water. Boil water advisories and Safe Drinking Water Act violations followed[213, 282]. Adverse health effects were reported by consumers and residents began protesting.

In September 2014, Flint resident LeeAnne Walters began documenting her experiences and family's health issues, which prompted her to begin archiving water samples and stop using the city water for cooking or drinking a few months later. Shortly thereafter, voluntary sampling by the city in response to growing complaints demonstrated that Walters' drinking water had high lead (104-397 ppb). Blood tests thereafter revealed that her child had elevated blood lead (i.e.,$\geq$ 5 µg/dL). In February 2015, a team led by Erin Brokovich took up the residents' cause, and even sent a letter to the Flint mayor and city council asserting the need for better corrosion control, but the letter was "ignored".[320] Around the same time, Walters and other residents began attending city council meetings to raise awareness,

but their complaints were still not addressed.[202]

Around February 2015 Walters reached out to technical experts such as Miguel Del Toral at EPA Region 5 and the Flint Water Department to decipher the cause(s) for her high lead levels. We became involved in April 2015 by assisting Del Toral and Walters in collecting a profile of samples from her tap just before the city replaced the city owned lead service line.[70, 258] Our results revealed extremely high lead (over 13,000 ppb) and the results became the basis for an ACLU documentary featuring the data and resident health concerns.[125, 141] In spite of these results and an associated memo from Del Toral that documented federal corrosion control law was not being followed and the "imminent and substantial endangerment" under which Flint residents lived,[130, 160] the Michigan Department of Environmental Quality (MDEQ) and the City of Flint repeatedly maintained the water was in compliance with all Federal standards through July 2015. Given that Del Toral's whistleblowing memo backed by our initial sampling data was dismissed by all the relevant authorities, a decision was made collectively by Virginia Tech, Curt Guyette at ACLU Michigan, and a group of residents to conduct an independent city-wide sampling event in early fall 2015, before water temperatures cooled and the corrosivity of the water was decreased.[125]

Sampling and Results: Virginia Tech funded the provision of 300 3-bottle sampling kits to test for lead and other metals. Guyette, Walters and a team of numerous other residents recruited participants via word of mouth and the internet, passed out, and collected kits at a local church in Flint, in collaboration with the Concerned Pastors for Social Action. The sampling instructions included a VT YouTube video demonstration link to aid residents in properly collecting their samples. Additionally, three days after kits were sent, a VT team traveled to Flint to assist with sample collection, and to conduct sampling of biological and other contaminants that required advanced training and sample preservation that was not

amenable to citizen science. Over the next four weeks, 277 VT lead kits were returned for analysis, and regular chlorine sampling was conducted in several events around the city in a collaboration between Walters and Virginia Tech.[285] The 90th percentile value from the city wide sampling event was 26.8 ppb, which was decisively over the 15 ppb lead action level.[259] Chlorine residuals in Ms. Walters' home were undetectable from August 17 to September 3.[108]

Results were distributed to residents through phone calls and letters, and summaries of the results were presented to the community by VT students and posted on our Flint Water Study website (www.flintwaterstudy.org), Facebook, and Twitter.[166] Based on the data, our team decided to issue a Do Not Drink advisory on our website in early September 2015, and participated in a press conference with residents contradicting authorities.[38] We also assisted a local pediatrician with an analysis of elevated blood-lead levels in Flint children, the results of which were presented at another press conference in late September 2015. We continued sampling for pathogens and shared results demonstrating high Legionella in large buildings and hospitals in late 2015[306].

Outcomes: On the basis of our findings and collaborations, the water supply was changed from the Flint River back to Detroit in October 2015. Our data on Legionella in large buildings eventually prompted a January 2016 public announcement about two Legionella outbreaks in 2014 and 2015 that caused at least 13 deaths[265, 306] and a federal emergency was declared the next day.[17] Eventually over a billion dollars of federal, state, and charitable relief and legal settlements flowed to Flint. There were at least three congressional hearings, and civil lawsuits and criminal trials for potentially responsible parties are ongoing. Additionally, the MI Lead and Copper rule was revised in 2018 to close compliance loopholes and incorporate additional requirements for sampling, lead service line replacement, and transparency of results.[282] Revisions to the federal LCR were announced in

December 2021 to take effect in 2024.[345] Several books about the crisis and its aftermath have been authored. Some of the ethical dilemmas that we encountered in exposing the crisis and instances of citizen science research misconduct, including falsification of results by improper sampling and putting lead sinkers into plumbing systems that inflated water lead measurements, are documented elsewhere.[281]

I.3.2 St Joseph, LA

Background: The St. Joseph, Louisiana (pop. 1,100) water system was constructed in the 1950's, but over time the town's agricultural economic base collapsed and the population decreased. A rapid decline in the quality of the town's water, which is sourced from two groundwater wells, was evident by the early 2010s. Between 2012 and 2016, at least 20 boil water advisories were issued, primarily due to water main breaks or equipment failures.[199] In 2013, iron levels were found to be as high as 9.74 ppm, 32 times higher than the EPA aesthetic standard (0.3 ppm).[368] While the Louisiana Department of Health (LDH) issued an Emergency Administrative Order in 2013 to remediate the water, the State Health Officer continued to insist to residents that their water was safe, as the iron is not a public health threat.[221]

By 2015, 21 Safe Drinking Water Act violations had been reported, including violations for coliform detections (six between 2014 and 2015), violations of public risk communication requirements (Public Notice Rule and Consumer Confidence Report Rule), and deficiencies in the water system's filtration system. In 2015 , the Louisiana State Legislature committed $6 million to fix the town's water infrastructure. Unfortunately, those funds could not be received until the Mayor released his financial records, which he refused to do.[191]

In early 2016, citizen water quality complaints, pictures and postings through social media

began reaching the media,[225] including the town's submission of a White House petition to declare a federal emergency which received 13,372 signatures for a town of less than 2000 people. However, the petition did not garner the required 100,000 signatures to warrant a mandatory White House response. As consumer complaints swelled, the LDH announced a partnership with the US EPA in February to investigate claims about St. Joseph's water system.[197] A separate investigative audit by the LA State Legislature released in March 2016 uncovered evidence implicating the Mayor in a scheme to misuse over $127,000 of municipal funds through the inappropriate awarding of construction contracts to family members.[266] In face of this continued controversy, residents reached out to Dr. Adrienne Katner at the LSU School of Public Health, who in turn reached out to VT to request additional support.

Sampling and Results: In March 2016, four 3-bottle sampling kits for lead and other metals were prepared at VT and sent through Dr. Katner to community partners in St. Joseph to collect samples from the community. Tests from 3 sites were returned, showing lead levels as high as 42 ppb and providing evidence of potential health dangers from the drinking water. Dr. Katner contacted LDH and the relevant residents to present and explain the results. The State took no immediate action, at which point, residents shared their results with two community organizations, The Green Army and the Louisiana Environmental Action Network. These groups presented the results to the State Legislature in an April 2016 hearing in support of a bill calling for state intervention to address St. Joseph's water quality issues.[126] Shortly after in June 2016, a fiscal administrator was appointed to St. Joseph, opening the door for outside financial assistance.[190]

Outcomes: Pressure by State Representatives on the LDH to investigate the results of VT and Dr. Katner prompted the state's Safe Drinking Water Program to test almost 100% (n=414) of the town's homes. Results revealed water lead in over 20% of homes

exceeded the lead Action Level of 15 ppb, with values up to 1810 ppb.[200] An official warning letter was issued by the LDH to the utility. A few months later in August 2016, a State Legislative Audit of the LDH's Safe Drinking Water Program highlighted oversight and enforcement issues,[267] including reliance on water systems for sample self-collection (calling integrity of samples into question), delays in sanitary surveys for active water systems, lack of follow-up to ensure significant deficiencies were resolved, and a lack of citations when public notifications for violations were not conducted. In December 2016 news of this situation expanded to the national media, including CNN[368] and NPR[152], and comparisons of St. Joseph's water system were being made to Flint, MI's. At that point LA Governor John Bel Edwards finally declared a Public Health Emergency.[318] Resident celebrations ensued as this order provided residents of the town with free bottled water for each household for four weeks.[196, 309]

By March 2017, $8M in upgrades to the water distribution system were underway,[85] and were followed by the passage of State House Bill 533 (Act No 263 effective August 2017)[159] to improve access to safe drinking water in St Joseph. Construction of the new system was completed in March 2018, providing over 75,000 feet of new water lines along with a new water treatment plant and municipal wells.[198] However, this solution did not solve all of St. Joseph's water problems, as in January of 2018, the LDH reported seven residual chlorine violations,[139] and consumer complaints did not entirely disappear. It was also immediately understood that state supported funding could not be applied to every water system in the state,[85] as over 400 of 1374 regulated water systems in the state also had violations of drinking water regulations.[51] Thus, the LA Rural Water Infrastructure Committee was created in 2019 to identify the most distressed water systems,[47] due in part to 1) issues uncovered in St. Joseph, 2) a State Legislative Auditor's report,[267] and 3) a drinking water infrastructure grade of D- received from the Louisiana Section of the American Society of

Civil Engineers.[39] It is estimated that up to $10 billion will be required to fix all systems in LA.[132]

I.3.3 Enterprise, LA

Background: In 2004, the unincorporated town of Enterprise, LA (pop. approx. 300) constructed a new water tower, which was not put into use for one year, allowing rust to accumulate inside the tank. Once the water tank was put into operation, rust began to seep into the water which was already high in naturally occurring iron and manganese, resulting in water quality complaints, frequent boil water advisories, and residents driving as much as 25 miles to wash clothes.[132] The Enterprise Water System had been under administrative order for Disinfection Byproducts (DBPs) Rule violations since 2014,[215] but the system had struggled with inadequate minimum residual chlorine violations since 2009, and bacterial violations since 2013. The Enterprise water board, after paying for the water tower and other system improvements, was approximately $500K in debt, which excluded the town from eligibility for state grants.[132] Thus, increasing water rates was the only remaining option to raise funds for further improvements. In March 2017, researchers from Tulane University collected microbial samples revealing the presence in drinking water of fecal indicator bacteria.[325] In September 2017, the water board, eager to find solutions, worked with the community groups Louisiana Environmental Action Network and the Council of Policy and Social Impact to request assistance from Dr. Adrienne Katner and the VT team, who had previously worked together in St. Joseph.

Sampling and Results: In September 2017, 3-bottle sampling kits for lead and other metals were prepared at VT and sent to community partners. As results began to come in, Dr. Katner traveled to Enterprise to collect additional samples for chlorine residual and

DBPs. Sampling revealed that 41% of the 17 homes sampled had lead above the 15 ppb action level, 71% exceeded the aesthetic standard for iron, and 94% exceeded the aesthetic standard for manganese. Additionally, 29% of 21 homes tested for chlorine had no detectable residual and 76% exceeded the MCL for TTHMs.[200] Dr. Katner contacted community partners, the water board, and affected residents to inform them of their results, along with factsheets to interpret them and personal communication when needed.

Outcomes: Upon consultation with the VT team, the water tower and operation of the treatment plant were identified as the most likely causes of poor water quality. With data to support resident suspicions, the water issues began receiving widespread media attention, which in turn triggered State oversight.[253] In 2018, one of the first actions of the newly formed LA Rural Water Infrastructure Committee (RWIC), which was created to address drinking water quality issues in rural parts of LA, was to identify the 10 most distressed water systems in the state which included Enterprise.[26] The RWIC recommended consolidation with nearby larger systems, a long-term process which got underway in 2020 when the nearby town of Harrisonburg, LA received a $1 million Community Development Block Grant to merge their system with Enterprise.[353] Continued requests for funding are being pursued to continue construction on the combined system and cover legal fees. In search of a more immediate solution, the town was able to partner with the VT team to conduct a study on real-world performance of point-of-use filters in summer 2019, providing free filters to selected residents during the study. Unfortunately the point-of-use filters clogged quickly in waters with high iron or other particulates, making them difficult to maintain and less cost effective than bottled water.[65, 173] Water issues have persisted, as the 2021 Enterprise Consumer Confidence Report (CCR) noted multiple violations of the MCL for THMs, chlorine levels up to 7.1 ppm, and exceedances of iron and manganese SMCLs. The 90th percentile lead value, however, was only 4 ppb.[34] To protect residents more sustainably over time, more

expensive water softening and treatment systems would be required, which are beyond what some residents in the area can afford.

I.3.4 Denmark, SC

Background: The City of Denmark, South Carolina (pop. 3,400) has long operated and maintained its own independent municipal water system, despite attempts to develop a county-wide water system to pool resources amongst Bamberg County (pop. 15,400) towns.[133] Denmark and the surrounding region have long faced high unemployment (10.8%) and low median incomes ($26,591),[81] and there have been financial concerns, exacerbated by mayoral corruption dating back to 2002.[167, 363] Water services were directly affected in 2006, when two of three active wells in the city were taken out of service following detection of dry cleaning solvents PCE and TCE above permissible levels.[364, 365] In response, the city developed plans to construct two new wells using a grant from the SC Chamber of Commerce, and in the meantime purchase water from Bamberg County.[134] Shortly after, a SC Department of Health and Environmental Control (SC DHEC) inspection in 2008 rated the overall water system as "Unsatisfactory."[102] Additionally, despite a 20% increase in water rates in July 2008,[293] multiple boil water advisories[135, 293] were issued in summer 2008, accompanied by continued complaints about discolored water,[212, 294, 295] accidents at the water treatment plant,[293] and the discovery of lead in one of three above ground storage tanks.[292]

In September 2010, the new wells were completed,[158] however this did not end the city's water troubles. Over the next several years, numerous city council meetings were dominated by complaints over water quality, water management, and costs.[102, 129, 157, 251] Additionally, the city would receive repeated "unsatisfactory" ratings from the SC DHEC in 2011

and 2012 leading to fines and the institution of a full-time city water manager.[103] Two concerned residents, Eugene Smith and Paula Brown, continued to document issues with water quality and health effects, and sought out additional support from VT after watching Dr. Edwards testify at a Congressional Hearing on the Flint Water Crisis on television.[251, 322]

Sampling and Results: In early 2017, 3-bottle sampling kits for lead and other metals, along with sampling instructions, were prepared at VT and sent to Mr. Smith and Ms. Brown, who recruited additional participants in their community. In subsequent months, samples from 54 homes were collected, in addition to intensive specialized sampling by the VT team in a fall 2017 visit (THMs, chlorine residuals and opportunistic pathogens). Iron and manganese were chronically higher than secondary EPA MCLs. This campaign revealed that more than 12% of first draw samples exceeded the 15 ppb standard, but since this was only slightly higher than the 10% allowed under the LCR, the cities' claim to be meeting the action level could not be challenged with statistical confidence. Even so, the system received continued citations for lack of appropriate testing and failure to properly maintain or address lingering issues with storage tanks.[243, 372] Sampling results were shared with all participating residents and the VT team traveled to Denmark to aid in interpretation and share full community scale findings. Results were shared with the community and media through a press conference in fall 2017.

Unable to prove a problem with known regulated contaminants, the VT team repeatedly asked the Denmark city government for permission to inspect and test the wells for possible bacterial contamination in early 2018. These requests were repeatedly accepted and then retracted, after our team was in transit.[322] Similarly, the VT team offered to conduct free testing to explore the use of polyphosphate to sequester the iron and manganese at the wellhead, but this testing was also refused.

Outcomes: After the VT team was not permitted to sample the wells, the city eventually

allowed researchers from University of South Carolina to accompany state officials to sample wells in August 2018. Pictures of that inspection were sent to Dr. Edwards, who noticed that one of the wells was being treated with HaloSan, a disinfectant registered as a pesticide, that is commonly used in pools and spas and is not approved by the EPA for potable water use.[131] Halosan can cause symptoms of skin and eye irritation and internal bleeding, which were generally consistent with resident symptoms reported to the VT team by residents.[290] Examination of state records show DHEC officials had approved of the use of HaloSan since 2008, despite a 2007 EPA report that classified the active chemicals in HaloSan as pesticides which pose health risks when ingested.[290] The time at which Halosan was dosed to water roughly corresponded to the onset of symptoms cited by Ms. Brown and Mr. Smith to us.

City officials blamed DHEC for the use of HaloSan, but continued to insist the water was safe after its use was stopped, despite a further $4,000 fine from DHEC in July 2019.[366] Additionally, two separate class action lawsuits against the city were filed by citizens concerned over potential health damages resulting from the 10 years of HaloSan use. In fall 2019, the city received $2.3 million in grants and loans to upgrade aging pipes[328] and hired a public relations team to attempt to repair damage that concerned residents had done, according to city officials, to Denmark's reputation.[366] Additionally, a bill that had passed through the SC state legislature in 2012 aimed at creating a regional sewer and water authority for Bamberg County remains shelved, primarily due to a continued lack of support from the mayor of Denmark.[296]

I.3.5 Chicago, Cicero, Berwyn, and Robbins, IL

Background: The Chicago Metropolitan Area has approximately 400,000 lead service lines,[68] more than any other US city, in part due to a mandatory plumbing code that

required new homes be connected to the distribution system using only lead pipes until the federal lead plumbing ban in 1986.[57, 146] The pervasiveness of lead plumbing and publicity of elevated water lead measurements[5, 73, 121, 144, 148, 156] have resulted in serious concerns over lead exposure from drinking water in Chicago and the suburbs that directly purchase treated surface water from Chicago.[84, 147]

In one such suburb, Cicero (pop. Approx. 80,000), concerns were heightened in 2017 when the mandatory annual consumer confidence report sent by the town noted their failure in monitoring pH and orthophosphate levels added to water to control corrosion.[332] Delia Barajas, co-founder of an environmental justice group Ixchel, reached out to VT with a desire to obtain water lead testing. In partnership with Ixchel, VT prepared and delivered 10, 3-bottle sampling kits to collect first draw, 1-minute flushed, and 3-minute flushed samples from 10 homes. This testing found 7/30 total samples with lead above 15 ppb, and a 90th percentile value of 19.9 ppb, which is above the EPA action level of 15 ppb. In contrast, Cicero's mandatory Lead and Copper Rule sampling conducted shortly thereafter in 30 homes, found 0 of 30 samples above 15 ppb and a 90th percentile value of just 6.4 ppb. In reviewing Cicero's water lead sampling protocol, Ms. Barajas, Ixchel co-founder Diane Johnson, and the VT team discovered the instructions allowed water use in the home, that could negate the required 6-hour minimum period of no water use. The next summer, Ixchel requested a broader sampling campaign to better characterize lead levels in the community.

Sampling and Results: In summer 2018, about 120 3-bottle sampling kits for lead and other metals, along with sampling instructions (in English and Spanish) were prepared at VT and delivered to Cicero and nearby Berwyn (pop. Approx. 55,000) by VT researchers, who assisted residents in sample collection. Participants were recruited via word of mouth by Ixchel representatives and VT graduate student Chivonne Battle, who visited community sites such as churches or salons to further engage the community. Overall, samples from 77

homes were collected in Cicero and Berwyn in August 2018, and an additional 68 samples in Robbins (pop. Approx. 5,000) one month later in September (Table J.1). In Cicero and Berwyn, communities with high known prevalence of lead plumbing, 34% of homes had "concerning" lead of at least one sample above 10 ppb or two of their three samples above 5 ppb. In Robbins, however, a community where only one home had a confirmed lead pipe, only 1.5% of homes exceed those criteria. Building momentum from these campaigns, Ms. Battle further built a close, working relationship with Reunite Chatham, a community organization focused on Chicago's South Side. Reunite Chatham and VT conducted a large sampling event in July 2019, reaching 194 homes, 29% of which were considered concerning. Analysis of first draw and flushed samples confirmed observations of others[84] and further showed short-term flushing instructions to decrease water lead levels to protect consumers, actually worsened lead in water and consumer exposure.

Sampling results were mailed to all participating residents, and community partners Ixchel and Reunite Chatham called residents with concerning lead levels to explain the results and offer remediation options to reduce water lead exposure. A baseline phone script provided by VT was also translated into Spanish and used to share results in the primarily Hispanic suburbs of Cicero and Berwyn. All findings and data were shared with EPA Region V throughout the sampling campaigns, and with the public and media at press conferences held at the Cicero Town Hall (August 2018) and Nat King Cole Park in Chicago (August 2019).

Outcomes: Results of sampling became front-page news in the Chicago Tribune,[145] and drew continued attention over time.[146, 147] Cicero was issued a water quality violation by the EPA Region V. In 2018, the Illinois EPA sent a series of violation notices to Berwyn for failing to submit lead testing data to the state in 2019, despite sending official notices that the water was safe, and various community meetings continued to spark debate over

water quality.[163, 279] Additionally, the City of Chicago is currently conducting a corrosion control study to determine potential options to reduce lead release in such communities and recommendations for flushing times were increased from 30 sec-2 min to 5 min to reduce lead levels based on testing results. Cicero has published 90th percentile lead values from 6.7 to 8.1 ppb from 2019-2021, with at least one home above the action level each year. Berwyn has published values between 3.6 and 8.9 ppb, with only one home above the action level in 2019.

I.3.6 Moore, OK

Background: The City of Moore (pop. 62,000) water source consists of groundwater from its own wells and potable surface water purchased from nearby Oklahoma City, which are blended in varying percentages over time to serve to residents. Some of Moore's wells are known to draw from the Central Oklahoma aquifer[69, 248] which has been reported to have high levels of naturally occurring arsenic. Compliance testing revealed arsenic levels exceeding the EPA MCL of 10 ppb in three separate wells across various official regulatory sampling events between March 2018 and 2019, which were not publicly revealed until a May 2019 notice.

From summer 2018 to spring 2019, drinking water samples were collected by residents who were concerned about arsenic and chromium levels, which revealed arsenic up to 14.7 ppb, and exceeding the arsenic MCL in 4 of 5 samples. The citizens issued a report to the owner of the sampled home, who discretely brought these results to the attention of Moore's City Council at a regular public meeting in April 2019. The homeowner was publicly thanked for "coming and presenting this instead of just posting something negative online".[337] Two days later, the city collected a voluntary sample from one of the wells with previous arsenic

issues, confirming that the arsenic was above the MCL. Subsequently, arsenic results from the concerned citizens dropped below the MCL. In May 2019, city officials notified the public of the original 2018 exceedances in one well, with a similar notice for a second exceedance in a different well sent out in October. A local tax was introduced in order to raise funds to rehabilitate the high arsenic wells, so that the town could eventually save money on purchases of OKC water with low arsenic.

In August 2020, the concerned citizens approached us with another sample they had collected above the arsenic MCL, prompting concern that the city was once again pumping from the high arsenic wells in order to save money. We agreed to support a citizen sampling campaign to investigate whether the Moore potable water was meeting Federal Regulations.

Sampling and Results: Sampling kits and instructions were prepared at VT and sent to the concerned citizens for weekly testing in fall 2019, followed by monthly testing thereafter until December 2019, where arsenic detections remained below the MCL (Figure I.1). Kits contained bottles for arsenic and other metals and hexavalent chromium, in addition to at-home kits for pH and chlorine residual. Residents collected all samples from their own homes. Additional VT sampling in fall 2020 documented that the arsenic MCL was exceeded from 9-2-2020 until 9-28-2020, and results were immediately shared with the residents. The concerned citizens conducted additional tests with an independent certified lab to confirm our result.

To respect the residents desire to remain anonymous, VT contacted city officials about the high arsenic results on 9-23-2020 via email. There was no response. When follow up emails were sent on 9-29, city officials dismissed us by stating "there is no issue with the water," and accusing us of being "combative" and seeking "fame" (See representative emails Section J.1). Within just 6 days of the initial response denying knowledge of high arsenic in the water, our on-going monitoring revealed that the arsenic levels plummeted from 15.4 to 0.6

ppb (Figure I.1). We speculate that an arsenic contaminated well that was being pumped to reduce the expense of purchasing Oklahoma City water was shut off, causing the immediate decline in arsenic.

A series of freedom of information act (FOIA) requests were submitted by VT, on behalf of the concerned citizens who still wished to remain anonymous for fear of retaliation, to Moore to better understand why wells with high arsenic were being used. Requests for emails, results of mandatory public well testing, and testing reports for some wells were provided. However, our requests for information about whether the wells with high arsenic that were supposed to be remediated were in use, causing the high arsenic, were inexplicably not provided to us (Section J.2-J.4). Our monthly monitoring continued to the time of this publication and confirmed that arsenic remains below the 10 ppb MCL.

Outcomes: In the aftermath of our sampling the OK DEQ independently identified "critical" issues with three Moore drinking water wells in site inspections in June 2020 and February 2021. A further inspection in September 2021 revealed an additional 8 issues, which were referred to the Water Quality division to request additional engineering support (Section J.5). Citations included sanitary issues such as rusted or inadequate well caps and the fact that only 3 out of 54 inspected well flow meters were operational. It seems plausible that the disabled flow meters are why Moore could not provide data as to whether the wells with high arsenic were being used.

State inspectors also independently flagged Veolia Water, the contractor responsible for managing Moore's water supply, for violations in several nearby towns.[61] In Heavener, OK (pop. 3,500) concerned residents and a state representative held community discussions in spring and summer 2019 regarding water quality concerns. In August 2019, Veolia officials denied problems with the water, alleging that attention was being drawn to such issues for "political gain".[219] A few days later in August 2019, a state inspection of the treatment

plant found multiple major violations, including errors in chemical additions and filter cleaning protocols and a lack of monitoring of water quality and flow.[66] In October 2019 after continued noncompliance, OK DEQ issued a $3,030,000 fine to Veolia.[25]

Several months later in March 2020, another treatment plant managed by Veolia in El Reno, OK (pop. 19,000) came under fire when an employee submitted a whistle-blower report alleging that a supervisor had directed them to illegally dilute samples prior to analysis. That supervisor was fired as a result, and his actions were characterized by Veolia as a misguided "lone-wolf former employee".[362] Veolia's problems in the region and the incident in Moore were the subject of a local news story.[61] We did not publicize our results because the arsenic levels plummeted after we drew the cities attention to the problem in September 2020.

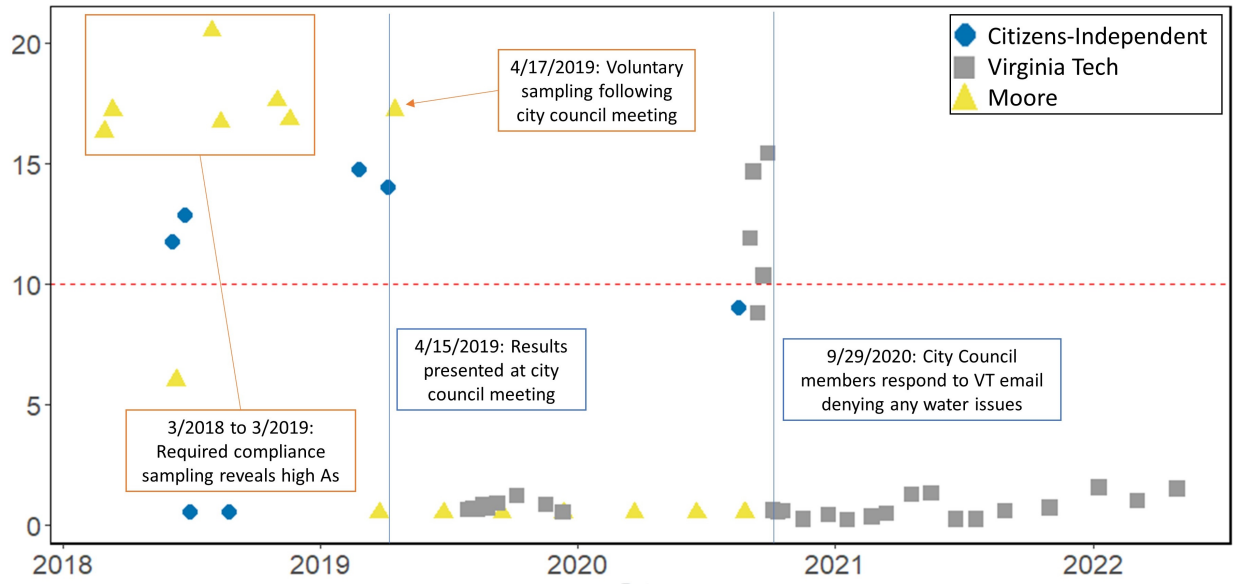


Figure I.1: **Summary of arsenic testing conducted by the City of Moore, Oklahoma Department of Environmental Quality, and VT**

I.3.7 Unnamed town in CA

Background: A resident in a CA town (pop. approximately 90,000) undertook a home renovation, including the installation of a new copper plumbing system, from 2014 to 2017. In such cases, elevated copper release is not uncommon prior to and during the early stages of formation of a protective scale, which can reduce corrosion over time.[114, 218, 348] However, in summer 2020, at least 3 years after their plumbing system had been completed and after increased water consumption from the home during COVID-19 stay-at-home orders, the resident's family noticed a green tint in their hair. In fall 2020, the resident used home test kits and had their water tested by a private laboratory, revealing copper above the EPA action level of 1.3 mg/L. In response to these concerns, alongside anecdotal complaints from other customers, the water utility conducted extra copper testing of 75 homes, revealing 10 above the action level. Following a re-test with a flushing protocol, however, only 2 remained above the action level, prompting the utility to conclude copper was an isolated problem in specific home plumbing systems. In response, in February 2021, the resident purchased a reverse-osmosis filter along with bottled water for drinking and worked with a private treatment company to install a soda ash feed system to increase the water pH. The resident, who had a PhD and a strong chemistry background, was still interested in monitoring the performance of this system and understanding why their pipes were experiencing continued corrosion so they reached out to the VT team in February 2021.

Sampling and Results: Sampling kits, including bottles for copper and other metals, and instructions were prepared at VT and sent to the resident in CA. At-home tests for copper and pH were also included. The resident collected three quasi-weekly samples from two locations in the home and sent the water back to VT for analysis. Sulfate reducing bacteria and chlorine residual were also tested for at selected sampling times. In parallel, the resident

and the water utility sent VT larger volumes of source water, which were used to conduct pipe segment cuprosolvency tests in the laboratory at VT under varying remediation options to further understand potential solutions. Results revealed copper frequently above the 1.3 mg/L EPA action level, especially when the pH amendment system was not being used, with levels reaching as high as 5.1 mg/L. Additionally, sporadic episodes of pH as low as 6.81 were documented in the source water and are believed to be associated with more copper corrosion and release in the home. Such pH variation is likely related to fluctuations in utility use of well vs. surface waters for their water supply. Sampling results were tabulated on a shared spreadsheet giving the resident real-time access, and trends were discussed on semi-regular video calls with additional calls/emails as needed. The resident has been solely responsible for further distribution of these results and has been in communication with the water utility and the private treatment company.

Outcomes: The utility is not legally required to take responsibility for copper issues in relatively new homes. Therefore, residents must manage these problems on their own. Compliance with the LCR creates a loophole for copper, as it is determined in monitoring homes built before 1986, and the city has been meeting the copper action level in these homes, with a 2022 90th percentile of 0.53 mg/L versus the action level of 1.3 mg/L. In an attempt to help this consumer, we evaluated granular activated carbon, anion exchange, reverse osmosis, and multiple levels of pH adjustment over a period of 9 months and none appear to provide a simple solution to the elevated copper. In the meantime, the resident continues to provide filtered water for their family and pets, especially after anecdotal claims that a dog died from consuming water with elevated copper.[367]

I.3.8 Unnamed town in South Carolina

Background: In late 2021, a resident contracted acanthamoeba keratitis, a rare eye infection that can occur from exposure to soil or water contaminated with free-living amoebae of the genus Acanthamoeba. Due to their concerns over drinking water as a possible source, the resident contacted the local water utility in January 2022 for potential testing and remediation measures. Acanthamoeba is not yet regulated under the Safe Drinking Water Act and, therefore, not part of routine monitoring by utility laboratories. The water utility, which serves over 150,000 consumers, wanted to understand any possible acanthamoeba sources in their distribution system but faced difficulty finding commercial laboratories that test for acanthamoeba. A search of the peer-reviewed literature led the utility to research articles on the topic from our team. We began collaborating after the utility reached out for assistance with acanthamoeba sampling.

Sampling and Results: Our team devised a rigorous sampling protocol and prepared an instruction sheet and a YouTube sampling video.[32] We shipped a sampling kit for the utility personnel to collect water and biofilm swab samples at the bathroom spigot, bathroom showerhead, and hose bib in the resident's house, and the finished water from the treatment plant. The utility team also took pH, chlorine and temperature readings using approved field methods at all locations. Sampling was conducted in February 2022 and the kit was shipped overnight to VT for analysis. Acanthamoeba were not detected at significant levels in any collected samples. Results were shared with the utility.

Outcomes: Due to the lack of regulations and standard procedures for detection and monitoring of acanthamoeba, determining an analytical method presented challenges. Our team evaluated two previously published protocols for the quantitative polymerase chain reaction (qPCR) analysis of Acanthamoeba castellanii.[269, 276] After months of troubleshooting the

assays, qPCR results indicated that A. castellanii was likely not present in any of samples, even though a result was above limit of detection in 1 of 3 replicates. Generally, 2 of 3 replicate samples must be obtained to report the presence of the pathogen. Droplet Digital PCR (ddPCR) was found to be a more definitive method of analysis. A. castellanii were not detected at any of the sampling locations using ddPCR. Therefore, the concerns of the utility over possible pathogen growth in the water supply were reduced by this sampling that they coordinated. The problems we experienced in assisting this consumer are expected given that methods of detecting these emerging problems with pathogens have not yet been standardized.

I.3.9 Harrisonburg, VA

Background: A 60+ year old man in Harrisonburg, VA (pop. 53,000) was hospitalized with COVID-19 in November 2021, and 18 days into recovery at the hospital was further diagnosed with Legionnaires Disease, a form of pneumonia caused by the bacteria Legionella pneumophila, and the leading cause of waterborne disease and deaths in the US.[80] While it was unclear if the infection was acquired at the hospital or elsewhere, the family desired water tests at the home. Ms. Faythe Silviera, a family member, contacted the local health department, the City of Harrisonburg, and the VT Water Quality Extension in 2021, before hiring a local lab that collected a showerhead sample, which tested positive for Legionella pneumophila. To mitigate Legionella growth, the family flushed their pipes and temporarily raised the water heater temperature to 150°F before reducing it to 140°F, after reviewing existing CDC guidelines.

Sampling and Results: Our team decided upon a sampling approach similar to the South Carolina town (see previous case study) and a co-author (SR) hand-delivered the kit and

explained the sampling protocol to Ms. Silveira in April 2022. She further reviewed the instruction sheet and YouTube video and collected water and biofilm swab samples at relevant locations in the bathroom, hose bib and water heater. Ms. Silveira also measured pH, temperature and total chlorine using a commercial aquarium pH color strip test, a thermometer and a field chlorine meter, respectively. All samples collected were negative for Legionella pneumophila possibly indicating effective remediation. A second water heater was sampled in the home using water and biofilm swab sampling and those samples were also negative. Results were shared electronically with Ms. Silveira.

Outcomes: The results suggested that the corrective measures taken based on the CDC advice may have been effective at mitigating Legionella within the system. The VT team engaged various students to coordinate sampling and conduct the tests and learn about the potential for the spread of Legionella in home plumbing systems as opposed to more commonly studied cases in larger systems like hospitals, hotels, or nursing homes.

	Flint	St. Joseph	Enterprise	Denmark	Chicago	Moore	California	South Carolina	Harrisonburg, VA
Initial Contact	April 2015	March 2016	September 2017	Early 2017	Summer 2018	June 2019	February 2021	January 2022	March 2022
End of Active Sampling	October 2019	Summer 2016	Fall 2019	August 2018	Fall 2019	Ongoing	Ongoing	July 2022	July 2022
Sampling Kits	Lead sampling kits	Lead sampling kits	Lead sampling kits	Lead sampling kits	Lead sampling kits	Arsenic testing kits	Copper testing kits	Microbiological sampling kits	Microbiological sampling kits
Who collected samples?	Residents with assistance from on-site VT team for microbiological sampling, pH, temperature, dissolved oxygen, DBPs and chlorine residuals	Residents collected samples from their own homes	Residents with assistance from on-site Dr. Katner for DBPs and chlorine residuals	Residents with assistance of on-site VT team for THMs and chlorine residuals	Residents with assistance of on-site VT team	Residents collected samples from their own homes	Two residents collected samples from their own home	Utility personnel measured pH, temperature, and chlorine residuals using field probes	Resident collected samples from their own home
Who recruited participants?	Concerned citizens and activists who formed Coalition for Clean Water and ACLU Michigan representatives	Concerned citizens	Louisana Environmental Action Network and Council of Policy and Social Impact representatives	Concerned citizens led by Paula Brown, Eugene Smith and Deanna Miller Berry	Ixchel representatives led by Delia Barajas and Diane Johnson and VT graduate student Chivonne Battle	Residents collected samples from affected homes in their community	Residents collected samples from affected homes in their community	Utility personnel collected samples from the affected home and water treatment plant	Resident collected samples from their home
Results	277 samples returned: 17% above lead action level 90th percentile value of 26.8 ppb	3 sites tested: Lead up to 42 ppb	17 homes tested for metals: 41% above lead action level 71% above iron SMCL 94% above manganese SMCL 21 homes tested by Dr. Katner: 29% had no Cl residual detected 76% above MCL for TTHMs	54 homes sampled: Iron and manganese "chronically" higher than SMCLs 12% of samples exceeded 15 ppb lead action level	Cicero and Berwyn: 77 homes tested for lead 34% had one sample > 10 ppb or two > 5 ppb South Side: 194 homes tested for lead 29% had one sample > 10 ppb or two > 5 ppb	Arsenic above MCL from 9-2 until 9-28 2020	Copper frequently above 1.3 ppm action level pH as low as 6.81 has been documented	Acanthamoeba were not detected at sufficient levels in any collected samples.	All samples were negative for Legionella pneumophila
How were results used?	Distributed to residents, presented to community and media at local press conference and online. Also utilized by pediatricians in blood lead press conference.	Dr. Katner contacted residents and LDH. Residents shared results with The Green Army and Louisiana Environmental Action Network, who brought them to LA State Legislature	Dr. Katner contacted community partners, the water board, and affected residents with their results	Distributed to residents and presented to the community in-person by the VT team.	Mailed to residents, community partners called. Shared with EPA Region V and with public/media at press conferences	Shared electronically with residents, but VT contacted city officials to share concern over arsenic.	Shared electronically with resident, who has been in contact with the utility and a private treatment company	Shared directly with the utility	Shared directly with concerned resident

Table I.1: **Summary of each case study.**

I.4 Discussion

The case studies described in this article span the past seven years (2015-2022). A range of results, time scales of involvement, regulatory actions taken, and lasting impacts on the water system were documented (Table I.1). Many of these changes are attributable to how the issue of drinking water quality and violations were elevated nationally following our work in Flint, MI and St. Joseph, LA, the resulting press coverage, and declaration of the water emergencies. Residents, who had been dealing with poor quality water for years prior, suddenly found their complaints addressed by state and local officials because of fears that a failure to act could create the next Flint.[85, 225] In addition to overhauls of the drinking water systems, both cases were also unique in that they changed the regulatory environment in the state, with the new state LCR in MI and the RWIC in LA.[23, 28]

Following these developments, the ability of collaborations between university researchers and citizen scientists to effectively challenge misinformation from authorities could not be questioned. However, government agencies also became more proactive because they simply could not afford to repeat these investments. Governor Bell Edwards in LA, after funding the infrastructure replacement in St. Joseph, even stated "The simple fact of the matter is we can't replicate this effort around the state because we don't have enough money."[85] The amounts of funding necessary to upgrade infrastructure throughout LA[215] and the nation[132, 225] are astronomical. Even national infrastructure investments, like the 2021 $1 trillion Infrastructure Bill H.R. 3684 were spurred at least in part by the decline in public trust and attention drawn to lead in drinking water in the aftermath of Flint. Even the $15B allocated for lead pipe replacement in this bill however, only represents about $53 per person consuming public drinking water[97], which pales in comparison to funding for system repairs and legal settlements in St. Joseph or Flint of approximately $7,200 and $15,000 per

person respectively.

Regulators also openly admitted to us that they were now more aggressive in issuing citations and more reluctant to give blanket claims that the water was safe, which might be falsified by testing of our team. We consider this to be a positive development. All of these changes also make it less likely that some of the more astonishing results of our citizen science sampling campaigns will have such dramatic impacts in the future.

At the same time, the funding and media coverage that our work helped mobilize in Flint, MI and St. Joseph, LA, created a model aspired to by residents in other beleaguered cities, who understandably wanted similar attention and relief for their own water woes. According to Goodhart's Law, "When a measure becomes a target, **it ceases to be a good measure**." We therefore have to temper the unrealistic expectations of our newer citizen science collaborators who openly aspire to become the next Flint or St. Joseph in terms of publicity and relief funding.

I.4.1 Citizen Objectives and Intervention Strategies

For purposes of synthesis, we demarcated the nine case studies into three chronological and consequential categories: 1) Declared public health emergencies (Flint, St. Joseph: 2015-16); 2) Chronic problems (Enterprise, Chicago, Denmark: 2017-present); and 3) Evolving citizen science concerns (Moore, CA, Harrisonburg, SC: 2019-present). The relative successes of each case can be evaluated in the context of national drinking water perception at the time they occurred.

In our work in Flint, MI and St. Joseph, LA, citizens were faced with urgent threats to their health, and our monitoring data strongly indicated that Federal drinking water standards were being violated in direct contradiction to official pronouncements the water

was safe. Our data and interpretation were subsequently verified independently. In Flint, our collaboration received national attention and the data and press coverage forced state and federal environmental agencies to respond and fulfill their regulatory responsibilities. Similarly, in St. Joseph, sampling results were brought to the LA state legislature, prompting confirmation sampling and drawing the attention of state agencies and the LA governor. Both campaigns created a national (and sometimes international) media sensation that drove aggressive responses. In both cases, the time between initial sampling data from the VT sample collection and declaration of water emergencies was less than one year.

In the aftermath of these citizen science successes, citizens in other areas also believed that they faced urgent public health threats, and a wide range of local organizations or coalitions of residents were again engaged. None of these cases resulted in declaration of water emergencies and state supplied bottled water was not forthcoming, although some communities such as Denmark eventually received private donations of bottled water. As a result, our work in Denmark, Chicago and Enterprise involved longer-term sampling efforts spanning more than 12 months. Additionally, intervention efforts shifted more towards providing specific outreach and advice to a community, because it was unclear if laws were broken and there was no evidence of state or federal agencies involved in cover-ups. Large-scale funding of infrastructure was not forthcoming.

In the much later cases, concerns of residents were in areas of unregulated and evolving science that do not involve violations of the SDWA. The purpose was to better understand the science of unregulated contaminants, with a long-term aspiration of improving policy or best practices and future regulations. The latter stages of work involved engagement and expertise of individuals with a large variety of expertise and specific needs. In CA, the concerned retired resident was a scientist who had goals of understanding the water chemistry relevant to elevated copper in their home. In Moore, the concerned residents

had also acquired what we considered to be graduate-level expertise. In these cases, the residents originated most of the research ideas and we merely assisted them in interpreting or confirming the results. In Moore, in fact, we were so dubious that the utility would pump water arsenic contaminated from wells after being caught by residents just months before, we figured our sampling would merely humor them and falsify their hypothesis. We were proven wrong. Residents in SC and Harrisonburg sought out our expertise and requested more guidance in how to proceed to better understand their system. In any case, the nature of our assistance evolved with the needs of our citizen science collaborators, as it does in relationships with academic scientists.

I.4.2 Outcomes post-Flint and St. Joseph

In Denmark, SC and Chicago, IL, the regulatory authorities issued citations to systems managing the water, but external funding was not made available for system upgrades. Results represented small versus major infrastructure upgrades. For example, while the government oversight was unchanged in Denmark, use of HaloSan was terminated and a small amount of funding was made available for infrastructure improvements. Similarly, provision of water filters provided a short-term improvement in safety of water used for cooking or drinking. Improvements in sampling and corrosion control studies in Chicago, and the consideration of system mergers and infrastructure improvements in Enterprise, LA, improved water safety. In SC, and Harrisonburg, VA, novel techniques of microbial monitoring and mitigation techniques were piloted and expected changes to policies or regulations are years in the future. In Moore, OK, solutions through sales taxes to treat water from problematic wells were already being pursued. In CA, a short-term treatment solution for copper contamination was achieved, however it is unlikely to result in significant near-term policy changes.

I.4.3 Reception from Water Authorities

When water quality issues are revealed independently by an academic team, it is understandable that parties responsible for management of the water system are rarely eager to cooperate, since the outcome might call into question their authority, judgment, and past pronouncements regarding water safety. In some cases, such as Moore, OK or Denmark, SC this included a refusal to allow outside sampling of wells or even share basic knowledge about whether wells contaminated with arsenic were used as water sources. On the other hand, ironically, when citizens reached out for assistance with emerging unregulated contaminants, cooperation with the authorities by utilities was offered graciously. This is partly a self-selection process, in that many progressive utilities actively seek assistance for customers to go beyond the SDWA, whereas many of our other engagements were with a select group of utilities breaking provisions of the SDWA. Academics engaging in such work should anticipate extremes of responses from cooperation to gentle pushback, and in some cases to denial of results and personal attacks.

I.4.4 Defining Success

In any collaboration between citizens and scientists, views of success are in the eye of the beholder. The perspectives of citizens and community groups can be diverse and complex. In Flint, MI, our initial data collection confirmed residents' suspicions, and we were initially given accolades. But during the national and international media coverage some groups believed we were given too much credit.[118, 186, 281] When our same data collection efforts showed improving water quality, some residents were concerned that it would draw the spotlight and funding away from the recovery, and expressed disapproval that we would present such data publicly without consulting them.[48, 101] When we exposed incidents of

citizen science misconduct, including lead fishing sinkers in plumbing of a resident seeking to raise money from gofundme due to "poisoned" water and in support of a lawsuit, many residents became angry with us and threatened a defamation suit.[178, 319] When billions of dollars and Hollywood movies are involved, such controversies may be unavoidable between activists dedicated to causes and scientists devoted to data analysis.[281]

From an academic perspective, citizen science is sometimes viewed as threatening funding from authorities, which caused some professors criticize our work exposing the Flint Water Crisis.[307] That criticism was not widely supported[241], although it did continue ongoing discussions over calls for guardrails and standardization of citizen science.[128, 242, 274, 275, 283] Additionally, some efforts required years of engagement and a refusal to give up as in the case of discovering dosing of Halosan to Denmark, SC water. Funding all of the work out of our own pocket or with occasional financial donations made to our team made such sustained efforts difficult to maintain.

Finally, most studies do not result in peer reviewed publications. Of the approximately 100 cases examined at citizens' requests, the vast majority found nothing of note. Even cases described in this paper rarely resulted in academic publications, as only Flint results were directly published, with tangential papers resulting from specific technical aspects of water issues in Enterprise and CA. Thus, most cases did not result in any measure of traditional academic success, in terms of publications and research funding. In the few which did, these were only achieved along with substantial controversy. In fact, other academic researchers often took insights from our open science blogs, data, ideas without appropriate attribution, because many journals discourage citation of non-peer reviewed sources. On the other hand, in all cases residents were empowered with scientifically sound information about their water quality. In some cases, varying degrees of lasting, meaningful changes were produced. For academics interested in pursuing such citizen science-based work, understanding the differing

opinions as to what constitutes "success," and facing dilemmas of sometimes conflicting loyalties to scientific data versus advocacy, are an inextricable part of such work.

Appendix J

Supporting Information for Appendix H

J.1 Email correspondence with Moore, OK city officials

From: Jerry Ihler <JIhler@cityofmoore.com>
Sent: Friday, September 25, 2020 10:56 AM
To: Robert Pistole
Subject: RE: High arsenic detected in Moore potable water.

Robert … Check with the City of Norman to see if they have received a similar email from this professor. I will forward your email to Randy so he is aware of it.

Jerry Ihler, PE
Assistant City Manager
City of Moore
301 N. Broadway
Moore, Oklahoma 73160
(405) 793-5200
jihler@cityofmoore.com

From: Pistole, Robert <robert.pistole@veolia.com>
Sent: Wednesday, September 23, 2020 3:04 PM
To: Jerry Ihler <JIhler@cityofmoore.com>
Subject: Fwd: High arsenic detected in Moore potable water.

FYI. Your thoughts?

---------- Forwarded message ---------
From: <edwardsm@vt.edu>
Date: Wed, 23 Sep 2020 at 11:56
Subject: High arsenic detected in Moore potable water.
To: <robert.pistole@veolia.com>

Hi Robert, I have been trying to call and email. The emails listed online do not work, nor does Mr. Eddy's email.

Please confirm receipt of this communication.

Best Regards,

Marc

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, September 23, 2020 12:36 PM
To: SEDDY@CITYOFMOORE.COM; Robert.Pistole@Vediawaterna.com
Subject: High arsenic detected in Moore potable water.

Dear Steve Eddy and Robert Pistole,

Allow me to introduce myself. I am Professor Marc Edwards of Virginia Tech (short bio attached), and we have been monitoring Moore potable water on and off for the last few years.

Recently, we have collected several potable water samples that exceed the 10 ppb arsenic MCL, and we have confirmed the legitimacy of our concern with a third party laboratory.

We would like to work proactively with you to figure out what is going wrong with the water supply, and to determine appropriate and timely steps for public notification of the potential health risks.

Can we arrange a zoom call to discuss possibilities, in hopes of working collaboratively with you to address this issue as soon as possible?

Alternatively, we could submit a FOIA for records on well use, but that would result in worrisome delays in notifying the public and regulatory agencies about this potential health concern.

Thank you in advance for your consideration.

Best Regards,

Marc

From: Jerry Ihler <JIhler@cityofmoore.com>
Sent: Friday, October 2, 2020 2:15 PM
To: Brooks Mitchell
Subject: FW: Attention: High arsenic detected in Moore potable water.

Brooks … Robert forwarded his response to your email. Odd that he sent it to everyone but you.

Jerry Ihler, PE
Assistant City Manager
City of Moore
301 N. Broadway
Moore, Oklahoma 73160
(405) 793-5200
jihler@cityofmoore.com

From: Robert Pistole <robert.pistole@veolia.com>
Sent: Friday, October 2, 2020 11:39 AM
To: Jerry Ihler <JIhler@cityofmoore.com>
Subject: Fwd: Attention: High arsenic detected in Moore potable water.

Robert Pistole
Project Manager
Moore, Oklahoma
(405)793-5080 Office
(405)627-1842 Cellular
(405)793-5095 Fax

Begin forwarded message:

> **From:** edwardsm@vt.edu
> **Date:** September 30, 2020 at 9:35:55 AM CDT
> **To:** Mark Hamm <marks.hamm@cox.net>
> **Cc:** robert.pistole@veolia.com, GLewis@cityofmoore.com, AWebb@cityofmoore.com, DMcKenzie@cityofmoore.com, MHamm@cityofmoore.com, MHunt@cityofmoore.com, JBlair@cityofmoore.com, LWilliams@cityofmoore.com
> **Subject: RE: Attention: High arsenic detected in Moore potable water.**
>
> If the wells in question are indeed offline, then you are presently using other wells, which have arsenic levels over the EPA MCL.

I had hoped that you would want to know that we discovered high arsenic in your potable water, as confirmed through both our lab testing and samples that were submitted to a third party lab, and that you would also want to inform your customers about these potentially harmful exposures. The logical step would be to turn off, or treat, the water containing high arsenic in the problematic wells.

I have now received an unambiguous message that my hopes were misplaced, and that it was not an accident my prior phone calls and emails on this issue were ignored.
I will cease in my efforts to proactively address this concern with anyone from the city.

Marc

From: Brooks Mitchell <BMitchell@cityofmoore.com>
Sent: Wednesday, September 30, 2020 10:15 AM
To: edwardsm@vt.edu
Subject: City of Moore

Mr. Edwards,

The wells that you reference in your email are still off line and are not scheduled to rehabilitated until 2021. We will comply with all Oklahoma Department of Environmental Quality guidelines. During this project if we need consultants we will look to qualified people within the State of Oklahoma or the Southwest Region. To allege that the City of Moore would endanger their citizens is shameful.

Brooks Mitchell
City Manager
City of Moore, OK
(405) 793-5200

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, September 30, 2020 9:25 AM
To: 'Mark Hamm' <marks.hamm@cox.net>
Cc: robert.pistole@veolia.com; GLewis@cityofmoore.com; AWebb@cityofmoore.com; DMcKenzie@cityofmoore.com; MHamm@cityofmoore.com; MHunt@cityofmoore.com; JBlair@cityofmoore.com; LWilliams@cityofmoore.com
Subject: RE: Attention: High arsenic detected in Moore potable water.

Adam,

It is well-known that Moore has a problem with high arsenic in some wells. This issue has been acknowledged in the press for years, including as recently as fall last year.

According to Robert Pistole, project manager for Moore, tests in each of the nine offline water wells in 2000 measured about 40 parts per billion. Pistole said the city decided to stop using those wells and instead buy more of their water from Oklahoma City. "It didn't benefit the tax-paying citizens of Moore to treat the arsenic in the wells when we could just purchase the water from Oklahoma City," Pistole said. "The technology to lower arsenic levels was much more expensive then, and the rates to buy water from Oklahoma City were lower." ….Mitchell said reactivating these nine water wells would

save the city about $700,000 per year, and could also work towards stabilizing the rates that Moore residents pay for their water.
https://www.mooreamerican.com/news/local_news/sales-tax-election-nov-12/article_5cd5a684-3637-5771-bf20-91ca09c927bc.html

I do not think that my attempts to discuss this issue proactively, makes me "look like a jerk and reflects juvenile behavior."

I think it is highly likely, that someone associated with the City of Moore, decided to use wells that have arsenic well above the EPA MCL in an effort to save the city money.
If that is true, and I think it is, I figured someone at the city would want to get to the bottom of it sooner rather than later.

Marc

From: Adam Webb <AWebb@cityofmoore.com>
Sent: Wednesday, September 30, 2020 7:45 AM
To: Marc Edwards <edwardsm@vt.edu>
Subject: Re: Attention: High arsenic detected in Moore potable water.

Marc,
Your emails seem to be combative and that your just looking for your new "time of fame" to feed your ego. If you're truly concerned about water, we can have that discussion. By your emails, however, you simply seem rude and looking for a fight. Call the media if you wish. Do not make threats and do not reply while falsely quoting Mark Hamm. It not only undermines you as a doctor and person of character, it just makes you look like a jerk and reflects juvenile behavior.

To our knowledge, there is no issue with the water. We have been in talks with the EPA regarding other water situations you have not mentioned, which seems to discredit you The EPA has never stated there is an issue with arsenic levels. We also have third party testing and post the results. If you find something outside of the EPA and third parties that concern you, please email us those concerns for our consideration. We would be more than happy to take a look. The safety of our citizens is important to us. Even to the point of entertaining a conversation with someone such as yourself, who makes a terrible first impression, through email, at least.

Have a good day and thanks for reaching out (if there's really an issue and you're not just looking for an argument to make the news).

Sent from my iPhone

From: Mark Hamm <marks.hamm@cox.net>
Sent: Tuesday, September 29, 2020 9:45 PM

To: Marc Edwards <edwardsm@vt.edu>
Cc: robert.pistole@veolia.com; GLewis@cityofmoore.com; AWebb@cityofmoore.com; DMcKenzie@cityofmoore.com; MHamm@cityofmoore.com; MHunt@cityofmoore.com; JBlair@cityofmoore.com; LWilliams@cityofmoore.com
Subject: Re: Attention: High arsenic detected in Moore potable water.

I'll forward you email to the City Manager, Brooks Mitchell.
Sent from my iPhone

> On Sep 29, 2020, at 20:33, Marc Edwards <edwardsm@vt.edu> wrote:
>
> So there is actually a plan to not speak to me, and to ignore my prior phone calls and emails about the high arsenic. Is that what you are saying?
>
> Sent from my iPhone
>
> > On Sep 29, 2020, at 9:15 PM, Mark Hamm <marks.hamm@cox.net> wrote:
> >
> > The city publishes all reports of the water quality to the citizens we serve. You are welcome to notify the media if you feel so inclined.
> >
> > Thank you
> >
> > Sent from my iPhone
> >
> > > On Sep 29, 2020, at 18:19, edwardsm@vt.edu wrote:
> > >
> > > Dear Mayor of Moore, OK and Council Members,
> > >
> > > I have been trying to communicate important concerns about detection of elevated arsenic in Moore drinking water for more than a week now.
> > >
> > > Attempts to contact appropriate city officials include phone calls, sending emails to incorrectly listed addresses that do not exist for a Steve Eddy and Robert Pistole, and I have also left phone messages that have gone unanswered.
> > > For your information, incorrect email addresses are listed here with SIDWIS (http://sdwis.deq.state.ok.us/DWW/JSP/WaterSystemDetail.jsp?tinwsys_is_number=716896&tinwsys_st_code=OK&wsnumber=OK2001412)
> > >
> > > I am now coming to you. Can you have please make sure that someone contacts me about this important issue as soon as possible?

If not, I will have no choice, but to immediately escalate this public health concern with your regulator, the U.S. EPA and the media.

I have attached a short bio for your consideration. Thank you in advance for your attention.

Marc Edwards
University Distinguished Professor
Virginia Tech

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, September 23, 2020 12:57 PM
To: robert.pistole@veolia.com
Subject: High arsenic detected in Moore potable water.

Hi Robert, I have been trying to call and email. The emails listed online do not work, nor does Mr. Eddy's email.

Please confirm receipt of this communication.

Best Regards,

Marc

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, September 23, 2020 12:36 PM
To: SEDDY@CITYOFMOORE.COM; Robert.Pistole@Vediawaterna.com
Subject: High arsenic detected in Moore potable water.

Dear Steve Eddy and Robert Pistole,

Allow me to introduce myself. I am Professor Marc Edwards of Virginia Tech (short bio attached), and we have been monitoring Moore potable water on and off for the last few years.

Recently, we have collected several potable water samples that exceed the 10 ppb arsenic MCL, and we have confirmed the legitimacy of our concern with a third party laboratory.

We would like to work proactively with you to figure out what is going wrong with the water supply, and to determine appropriate and timely steps for public notification of the potential health risks.
Can we arrange a zoom call to discuss possibilities, in hopes of working collaboratively with you to address this issue as soon as possible?

Alternatively, we could submit a FOIA for records on well use, but that would result in worrisome delays in notifying the public and regulatory agencies about this potential health concern.

Thank you in advance for your consideration.

Best Regards,

Marc
<bioshort.doc>

From: Adam Webb <AWebb@cityofmoore.com>
Sent: Wednesday, September 30, 2020 7:49 AM
To: Mark Hamm
Subject: Fwd: Attention: High arsenic detected in Moore potable water.

My response. This guy just wants to be in the news again.

Sent from my iPhone

Begin forwarded message:

> **From:** Adam Webb <AWebb@cityofmoore.com>
> **Date:** September 30, 2020 at 6:44:43 AM CDT
> **To:** Marc Edwards <edwardsm@vt.edu>
> **Subject: Re: Attention: High arsenic detected in Moore potable water.**
>
> Marc,
> Your emails seem to be combative and that your just looking for your new "time of fame" to feed your ego. If you're truly concerned about water, we can have that discussion. By your emails, however, you simply seem rude and looking for a fight. Call the media if you wish. Do not make threats and do not reply while falsely quoting Mark Hamm. It not only undermines you as a doctor and person of character, it just makes you look like a jerk and reflects juvenile behavior.
>
> To our knowledge, there is no issue with the water. We have been in talks with the EPA regarding other water situations you have not mentioned, which seems to discredit you The EPA has never stated there is an issue with arsenic levels. We also have third party testing and post the results. If you find something outside of the EPA and third parties that concern you, please email us those concerns for our consideration. We would be more than happy to take a look. The safety of our citizens is important to us. Even to the point of entertaining a conversation with someone such as yourself, who makes a terrible first impression, through email, at least.
>
> Have a good day and thanks for reaching out (if there's really an issue and you're not just looking for an argument to make the news).
>
> Sent from my iPhone
>
> > On Sep 29, 2020, at 8:33 PM, Marc Edwards <edwardsm@vt.edu> wrote:
> >
> > So there is actually a plan to not speak to me, and to ignore my prior phone calls and emails about the high arsenic. Is that what you are saying?
> >
> > Sent from my iPhone
> >
> > > On Sep 29, 2020, at 9:15 PM, Mark Hamm <marks.hamm@cox.net> wrote:

The city publishes all reports of the water quality to the citizens we serve. You are welcome to notify the media if you feel so inclined.

Thank you

Sent from my iPhone

> On Sep 29, 2020, at 18:19, edwardsm@vt.edu wrote:
>
> Dear Mayor of Moore, OK and Council Members,
>
> I have been trying to communicate important concerns about detection of elevated arsenic in Moore drinking water for more than a week now.
>
> Attempts to contact appropriate city officials include phone calls, sending emails to incorrectly listed addresses that do not exist for a Steve Eddy and Robert Pistole, and I have also left phone messages that have gone unanswered.
> For your information, incorrect email addresses are listed here with SIDWIS (http://sdwis.deq.state.ok.us/DWW/JSP/WaterSystemDetail.jsp?tinwsys_is_number=716896&tinwsys_st_code=OK&wsnumber=OK2001412)
>
> I am now coming to you. Can you have please make sure that someone contacts me about this important issue as soon as possible?
>
> If not, I will have no choice, but to immediately escalate this public health concern with your regulator, the U.S. EPA and the media.
>
> I have attached a short bio for your consideration. Thank you in advance for your attention.
>
> Marc Edwards
> University Distinguished Professor
> Virginia Tech
>
> ---
>
> **From:** edwardsm@vt.edu <edwardsm@vt.edu>
> **Sent:** Wednesday, September 23, 2020 12:57 PM
> **To:** robert.pistole@veolia.com
> **Subject:** High arsenic detected in Moore potable water.
>
> Hi Robert, I have been trying to call and email. The emails listed online do not work, nor does Mr. Eddy's email.

Please confirm receipt of this communication.

Best Regards,

Marc

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, September 23, 2020 12:36 PM
To: SEDDY@CITYOFMOORE.COM; Robert.Pistole@Vediawaterna.com
Subject: High arsenic detected in Moore potable water.

Dear Steve Eddy and Robert Pistole,

Allow me to introduce myself. I am Professor Marc Edwards of Virginia Tech (short bio attached), and we have been monitoring Moore potable water on and off for the last few years.

Recently, we have collected several potable water samples that exceed the 10 ppb arsenic MCL, and we have confirmed the legitimacy of our concern with a third party laboratory.

We would like to work proactively with you to figure out what is going wrong with the water supply, and to determine appropriate and timely steps for public notification of the potential health risks.
Can we arrange a zoom call to discuss possibilities, in hopes of working collaboratively with you to address this issue as soon as possible?

Alternatively, we could submit a FOIA for records on well use, but that would result in worrisome delays in notifying the public and regulatory agencies about this potential health concern.

Thank you in advance for your consideration.

Best Regards,

Marc
<bioshort.doc>

From: Brooks Mitchell <BMitchell@cityofmoore.com>
Sent: Wednesday, September 30, 2020 10:08 AM
To: Adam Webb; Adam Webb; Danielle N Mckenzie; Danielle McKenzie; Glenn Lewis; Jason Blair; jason blair (contact); Louie Williams; Mark Hamm; mark hamm (contact); Melissa Hunt
Subject: Arsenic levels

Mayor and Council,

This is an obvious shakedown. We currently have no DEQ compliance issues. We test pursuant to state law and DEQ requirements. The rehabilitation program for the wells that are off-line will not start until April 2021. This program will be funded by the 1/8 cent sales tax. We did not begin collecting this tax until June 2020. We will comply will all DEQ requirements during this project. What Mr. Edwards thinks he knows is wrong.

Brooks Mitchell
City Manager
City of Moore, OK
(405) 793-5200

From: Adam Webb <AWebb@cityofmoore.com>
Sent: Wednesday, September 30, 2020 10:11 AM
To: Brooks Mitchell
Cc: Adam Webb; Danielle N Mckenzie; Danielle McKenzie; Glenn Lewis; Jason Blair; jason blair (contact); Louie Williams; Mark Hamm; mark hamm (contact); Melissa Hunt
Subject: Re: Arsenic levels

Thanks Brooks! Here's an article of note on Mr. Edward's.

https://www.detroitnews.com/story/news/michigan/flint-water-crisis/2019/04/26/hero-pariah-flint-water-expert-mark-edwards-fights-for-his-reputation/3546987002/

Sent from my iPhone

> On Sep 30, 2020, at 9:08 AM, Brooks Mitchell <BMitchell@cityofmoore.com> wrote:
>
> Mayor and Council,
>
> This is an obvious shakedown. We currently have no DEQ compliance issues. We test pursuant to state law and DEQ requirements. The rehabilitation program for the wells that are off-line will not start until April 2021. This program will be funded by the 1/8 cent sales tax. We did not begin collecting this tax until June 2020. We will comply will all DEQ requirements during this project. What Mr. Edwards thinks he knows is wrong.
>
> Brooks Mitchell
> City Manager
> City of Moore, OK
> (405) 793-5200

J.2 Summary of correspondence of 1st FOIA request in Moore, OK

VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: FOIA request

edwardsm@vt.edu <edwardsm@vt.edu> Wed, Sep 30, 2020 at 3:21 PM
[redacted]
Cc: Kory Wait <korydw@vt.edu>

Could be. I have submitted a revised request.

Marc

From: [redacted]
Sent: Wednesday, September 30, 2020 3:17 PM
To: edwardsm@vt.edu
Cc: Kory Wait <korydw@vt.edu>
Subject: Re: FOIA request

Do you want anything that shows cost per gal that OKC is charging? Maybe they are increasing prices? OKC is a very big part of Moore's problem.

> On Sep 30, 2020, at 1:21 PM, edwardsm@vt.edu wrote:
>
> Am I missing anything?
>
> ---
>
> **From:** edwardsm@vt.edu <edwardsm@vt.edu>
> **Sent:** Wednesday, September 30, 2020 11:02 AM
> **To:** cityclerk@cityofmoore.com
> **Subject:** FOIA request
>
> We have detected high arsenic in Moore drinking water. Given that the city refuses to discuss this concern with me, to better understand the cause(s) of the high arsenic, I am requesting the following under the Freedom of Information Act.
>
> 1. Records regarding volume of water draw from individual city wells from 2015 to present, and records indicating the levels of arsenic detected in each well.
> 2. Records that would allow me to determine the percentage (or volumes) of water purchased from Oklahoma City.
> 3. Documents (emails, reports, minutes of meetings, etc) of the mayor, city council members, city manager and/or robert.pistole@veolia.com, from October 1, 2019 to the present, on the subject of:
>
>   a. levels of arsenic in Moore well water
>   b. levels of arsenic in Moore potable water

[redacted] 1/2

c. costs of purchasing water from Oklahoma City, or discussions about the volume of water used from city wells
d. costs of treating the water for arsenic

4. Emails of the mayor, city council members, city manager and/or Robert Pistole from 9/21/2020-present, in reference to my attempts to communicate concerns about high levels of arsenic in Moore's potable water.

Thank you in advance for considering this request.

Marc

[redacted]

VIRGINIA TECH

Kory Wait <korydw@vt.edu>

It is coming....supposedly.

edwardsm@vt.edu <edwardsm@vt.edu> Thu, Feb 11, 2021 at 10:30 AM
To: Kory Wait <korydw@vt.edu>, Ishi Keenum <ikeenum@vt.edu>, [redacted]
Amy Pruden <apruden@vt.edu>

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Thursday, February 11, 2021 9:52 AM
To: edwardsm@vt.edu
Subject: RE: FOIA

It is on a flash drive. I will get it out in today's mail.

Thanks!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Thursday, February 11, 2021 8:50 AM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: FOIA

The clerk said they could email the file to this address.

If that is not possible, my mailing address is:

Marc Edwards

1623 Jefferson Forest Lane

Blacksburg, VA 20406

Best Regards,

Marc

[redacted] 1/3

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Thursday, February 11, 2021 9:32 AM
To: edwardsm@vt.edu
Subject: RE: FOIA

I still need a mailing address to mail your reports to you.

Thanks!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, February 10, 2021 12:17 PM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Indeed. 540 320 8740

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Wednesday, February 10, 2021 1:15 PM
To: edwardsm@vt.edu
Subject: RE: FOIA

May I have a phone number and I can have a cashier call you for payment?

Thanks!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, February 10, 2021 12:12 PM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Dear Jamie,

Thank you. My preference is to pay by credit card on the phone or by a website. If that is possible, may I have a phone number or an address?

If that is not possible, please tell me to whom I should make out the check, the total amount, and the exact address I should send it to in order to expedite delivery.

Best Regards,

Marc

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Thursday, February 11, 2021 9:32 AM
To: edwardsm@vt.edu
Subject: RE: FOIA

I still need a mailing address to mail your reports to you.

Thanks!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, February 10, 2021 12:17 PM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Indeed. 540 320 8740

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Wednesday, February 10, 2021 1:15 PM
To: edwardsm@vt.edu
Subject: RE: FOIA

May I have a phone number and I can have a cashier call you for payment?

Thanks!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, February 10, 2021 12:12 PM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Dear Jamie,

Thank you. My preference is to pay by credit card on the phone or by a website. If that is possible, may I have a phone number or an address?

If that is not possible, please tell me to whom I should make out the check, the total amount, and the exact address I should send it to in order to expedite delivery.

Best Regards,

Marc

[redacted] 2/3

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Wednesday, February 10, 2021 1:03 PM
To: edwardsm@vt.edu
Subject: RE: FOIA

Your request has been completed. It has a $25 research and flash drive fee that needs to be paid before we can release the information.

Thanks!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, February 10, 2021 8:16 AM
To: city clerk <cityclerk@cityofmoore.com>
Subject: FOIA

Dear City Clerk,

On September 30th, 2021, I made a FOIA request for certain records related to high arsenic in Moore water. You confirmed receipt of my request.

On December 2nd I asked for an update, and I have received no reply. In fact, other than responding one time to confirm that you received my FOIA request, I have not received any information from you at all. It is now more than 4 months since my September 30th request.

Can you please give me an update on the status of my FOIA request?

Best Regards,

Marc

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, December 2, 2020 2:33 PM
To: cityclerk@cityofmoore.com
Subject: FOIA

Hello,

Do you have any update, as to the status of my FOIA request for records on the decision to use well water with high arsenic?

Best Regards,
Marc

J.3 Summary of correspondence of 2nd FOIA request in Moore, OK

VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: Second FOIA Request

edwardsm@vt.edu <edwardsm@vt.edu> Wed, Mar 31, 2021 at 9:14 AM
To: Kory Wait <korydw@vt.edu>, [redacted]

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, March 31, 2021 9:14 AM
To: 'Jamie Weaver' <JWeaver@cityofmoore.com>
Subject: RE: Second FOIA Request

Dear Jamie,

I received the FOIA documents in a letter. Thank you.

I noticed that only information from 2018 was provided in what was sent. Is there a reason for not providing the requested documents from 2019 and 2020?

Also, we were told that the city has access to pdf's of the full report including chain of custody.

Hence, we are not sure why the city is going to the lab and paying them to produce hard copies, of electronic reports they already have.

Thanks in advance for answering these questions about the partial production of responsive documents.

We would prefer the documents in electronic form.

Best Regards,

Marc

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Wednesday, March 24, 2021 2:22 PM
To: edwardsm@vt.edu
Subject: RE: Second FOIA Request

Ok, we are working on this request. Thanks!

[redacted] 1/8

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Tuesday, March 9, 2021 5:34 PM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: Second FOIA Request

Hi Jamie,

Thank you. We are aware of that. We would like to see the original reports with chain of custody signatures.

Those are not on the website.

Marc

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Tuesday, March 9, 2021 5:08 PM
To: edwardsm@vt.edu
Subject: RE: Second FOIA Request

Hello-

Everything that you are looking for can be located in the link provided.

There is no charge for this information.

http://sdwis.deq.state.ok.us/DWW/JSP/WaterSystemDetail.jsp?tinwsys_is_number=716896&
tinwsys_st_code=OK&wsnumber=OK2001412

Thank you.

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Thursday, March 4, 2021 12:55 PM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: Second FOIA Request

Hello Jamie,

Thank you for mailing us the emails and documentation. We have an additional request that should be easier to fill.

[redacted] 2/8

We are requesting the complete water test analysis report results from Accurate Labs, for the roughly 57 water samples that are listed in the following 3 tables. These are samples collected in 2018, 2019 and 2020. It is our expectation that a pdf of the complete original lab report can be sent to us.

Thank you,

Marc

Lab Sample No.	Collection Date & Time	Sampling Point	Sample Location	Laboratory
CL28014-09	12/27/2020 10:30:00	WL018	WELL 19	ACCURATE, INC.
CL28014-10	12/27/2020 8:50:00	CL037	CL2 STATION-WELL 37	ACCURATE, INC.
CL28014-11	12/27/2020 7:10:00	WL011	WELL 11	ACCURATE, INC.
CL28014-12	12/27/2020 7:30:00	CL009	CL2 STATION - WELL 9	ACCURATE, INC.
CL28014-20	12/27/2020 11:00:00	WL021	WELL 22	ACCURATE, INC.
CL28014-24	12/27/2020 8:35:00	WL023	WELL 24	ACCURATE, INC.
CH27125-01	8/26/2020 14:40:00	WL018	WELL 19	ACCURATE, INC.
CH27125-02	8/26/2020 14:30:00	WL023	WELL 24	ACCURATE, INC.
CF19012-01	6/18/2020 14:00:00	WL018	WELL 19	ACCURATE, INC.
CF19012-02	6/18/2020 13:40:00	WL023	WELL 24	ACCURATE, INC.
CC24079-01	3/22/2020 11:40:00	WL018	WELL 19	ACCURATE, INC.
CC24079-02	3/22/2020 11:20:00	WL023	WELL 24	ACCURATE, INC.
CC24079-03	3/22/2020 11:00:00	WL012	WELL 12	ACCURATE, INC.
CC24079-04	3/22/2020 12:10:00	WL028	WELL 29	ACCURATE, INC.
CC24079-05	3/22/2020 11:30:00	WL029	WELL 30	ACCURATE, INC.

[redacted] 3/8

Lab Sample No.	Collection Date & Time	Sampling Point	Sample Location	Laboratory
BL20006-01	12/19/2019 11:39:00	CL041	CL2 STATION-WELL 41	ACCURATE, INC.
BL20006-02	12/19/2019 11:30:00	CL040	CL2 STATION-WELL 40	ACCURATE, INC.
BL20006-03	12/19/2019 11:20:00	CL039	CL2 STATION-WELL 39	ACCURATE, INC.
BL20006-05	12/19/2019 9:45:00	CL028	CL2 STATION-WELL 28	ACCURATE, INC.
BL20006-06	12/19/2019 9:15:00	WL001	WELL 1	ACCURATE, INC.
BL20006-07	12/19/2019 9:30:00	WL003	WELL 3	ACCURATE, INC.
BL20006-08	12/19/2019 10:15:00	WL007	WELL 7	ACCURATE, INC.
BL20007-01	12/19/2019 10:00:00	WL008	WELL 8	ACCURATE, INC.
BL20007-02	12/19/2019 10:40:00	WL016	WELL 17	ACCURATE, INC.
BL20007-05	12/19/2019 11:10:00	WL021	WELL 22	ACCURATE, INC.
BL13011-01	12/12/2019 13:45:00	WL018	WELL 19	ACCURATE, INC.
BL13011-02	12/12/2019 13:00:00	WL023	WELL 24	ACCURATE, INC.
BI17137-01	9/16/2019 14:30:00	WL018	WELL 19	ACCURATE, INC.
BI17137-02	9/16/2019 14:10:00	WL023	WELL 24	ACCURATE, INC.
BF26016-01	6/25/2019 14:40:00	WL023	WELL 24	ACCURATE, INC.
BF26016-02	6/25/2019 14:50:00	WL018	WELL 19	ACCURATE, INC.
BD19007-01	4/17/2019 10:05:00	WL023	WELL 24	ACCURATE, INC.
BD16115-02	4/14/2019 12:19:00	WL022	WELL 23	ACCURATE, INC.
BC27112-01	3/27/2019 10:10:00	CL009	CL2 STATION - WELL 9	ACCURATE, INC.
BC27112-02	3/27/2019 10:05:00	CL002	CL2 STATION - WELL 2	ACCURATE, INC.
BC27112-03	3/27/2019 9:55:00	WL010	WELL 10	ACCURATE, INC.
BC27112-04	3/27/2019 10:35:00	CL037	CL2 STATION-WELL 37	ACCURATE, INC.
BC27112-05	3/27/2019 10:25:00	CL038	CL2 STATION-WELL 38	ACCURATE, INC.
BC26100-01	3/26/2019 13:30:00	WL018	WELL 19	ACCURATE, INC.
BC26100-02	3/26/2019 13:50:00	WL023	WELL 24	ACCURATE, INC.
BC26100-04	3/26/2019 14:30:00	WL012	WELL 12	ACCURATE, INC.
BC26100-06	3/26/2019 13:40:00	WL029	WELL 30	ACCURATE, INC.

[redacted] 4/8

Lab Sample No.	Collection Date & Time	Sampling Point	Sample Location	Laboratory
AK20110-01	11/19/2018 9:40:00	WL029	WELL 30	ACCURATE, INC.
AK20110-02	11/19/2018 9:30:00	WL018	WELL 19	ACCURATE, INC.
AK20110-03	11/19/2018 9:50:00	WL023	WELL 24	ACCURATE, INC.
AH14228-01	8/13/2018 14:15:00	WL029	WELL 30	ACCURATE, INC.
AH14228-02	8/13/2018 13:40:00	WL018	WELL 19	ACCURATE, INC.
AH14228-03	8/13/2018 14:00:00	WL023	WELL 24	ACCURATE, INC.
AF12161-01	6/11/2018 13:50:00	WL018	WELL 19	ACCURATE, INC.
AF12163-06	6/11/2018 15:40:00	WL029	WELL 30	ACCURATE, INC.
AC29087-02	3/29/2018 10:00:00	WL023	WELL 24	ACCURATE, INC.
AC29087-08	3/29/2018 9:35:00	CL002	CL2 STATION - WELL 2	ACCURATE, INC.
AC29087-01	3/28/2018 9:15:00	WL012	WELL 12	ACCURATE, INC.
AC13155-01	3/13/2018 14:25:00	WL029	WELL 30	ACCURATE, INC.
AC13155-02	3/13/2018 15:00:00	WL018	WELL 19	ACCURATE, INC.
AC13155-03	3/13/2018 13:10:00	WL021	WELL 22	ACCURATE, INC.
AC13155-04	3/13/2018 13:20:00	WL011	WELL 11	ACCURATE, INC.

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Thursday, February 11, 2021 9:52 AM
To: edwardsm@vt.edu
Subject: RE: FOIA

It is on a flash drive. I will get it out in today's mail.

Thanks!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Thursday, February 11, 2021 8:50 AM

[redacted] 5/8

VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: Records Invoice.pdf

edwardsm@vt.edu <edwardsm@vt.edu>
To: Kory Wait <korydw@vt.edu>[redacted]

Wed, Mar 24, 2021 at 5:49 PM

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Wednesday, March 24, 2021 5:49 PM
To: edwardsm@vt.edu
Subject: RE: Records Invoice.pdf

Ok, I will mail your reports out tomorrow. Thank you!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, March 24, 2021 4:45 PM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: Records Invoice.pdf

Thanks. I have paid for it. Marc

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Wednesday, March 24, 2021 5:26 PM
To: edwardsm@vt.edu
Subject: Records Invoice.pdf

Hello Marc,

Your reports have been gathered and are ready to be mailed to you once the research fee has been paid.

The total of the invoice is $146.50 and is attached to this email.

Please contact 405-793-5032 to pay a City of Moore Customer Service Rep.

Thank you.

Jamie Weaver

[redacted] 1/1

VIRGINIA TECH

Kory Wait <korydw@vt.edu>

Re: Second FOIA Request

[redacted] Thu, Apr 22, 2021 at 1:20 PM

To: edwardsm@vt.edu
Cc: Kory Wait <korydw@vt.edu>

Again, it's taking them entirely too long to produce a few measly records. I know it's not two weeks yet, but still....

On Apr 12, 2021, at 3:09 PM, edwardsm@vt.edu wrote:

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Monday, April 12, 2021 2:18 PM
To: 'Jamie Weaver' <JWeaver@cityofmoore.com>
Subject: RE: Second FOIA Request

Hi Jamie,

Thanks so much for the FOIA documents. We really appreciate it.

Please note that there are three items that we initially requested (and paid for) that are still missing.

1. we are asking for chain of custody's, associated with the following sample log numbers from Accurate Labs:

BL20006

BL20007

BL13011

BI17137

BF26016

BD19007

BD16115

BC27112

CF19012

CH27125

CL28014

[redacted] 1/10

2. please also provide the full lab report, including Chain of Custody, for sample log # BL20006. This is missing from the provided documents.
3. We also requested the entire original lab report (Cover Page, Results, QC data and Chain of Custody) for the arsenic test of WL023 (Well 24) for the water sample taken on 4/17/19 that was indicated as "not use for compliance" in your responsive records provided to our first FOIA request.

Can you check into this?

Thank you!

Marc

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Tuesday, April 6, 2021 12:31 PM
To: edwardsm@vt.edu
Subject: RE: Second FOIA Request

Hello-

I just sent you 2019 & 2020 in today's mail.

Thanks!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, March 31, 2021 9:30 AM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: Second FOIA Request

Thanks.

Same address as before.

Marc Edwards

1623 Jefferson Forest Lane

Blacksburg, VA 24060

[redacted] 2/10

Marc

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Wednesday, March 31, 2021 10:29 AM
To: edwardsm@vt.edu
Subject: RE: Second FOIA Request

We looked over the requested documents and found the dates requested are missing.

We will send you the documents for 2019 & 2020 in the mail.

This will be at no cost. I will email you once I have the documents in hand.

Please advise a good mailing address.

Thanks!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Wednesday, March 31, 2021 8:14 AM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: Second FOIA Request

Dear Jamie,

I received the FOIA documents in a letter. Thank you.

I noticed that only information from 2018 was provided in what was sent. Is there a reason for not providing the requested documents from 2019 and 2020?

Also, we were told that the city has access to pdf's of the full report including chain of custody.

Hence, we are not sure why the city is going to the lab and paying them to produce hard copies, of electronic reports they already have.

Thanks in advance for answering these questions about the partial production of responsive documents.

We would prefer the documents in electronic form.

Best Regards,

[redacted] 3/10

VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: FOIA

edwardsm@vt.edu <edwardsm@vt.edu> Mon, May 10, 2021 at 5:04 PM

[redacted]

This exchange just got a bit weirder.

The “in our possession” thing is a famous FOIA dodge.

Marc

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Monday, May 10, 2021 5:03 PM
To: 'Jamie Weaver' <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Hi Jamie,

I gave you a list of analysis that were not delivered. Should I send it again?

We were told by the lab that you have those.

Also, when you said “him,” do you mean me?

I am confused.

Best Regards,

Marc

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Monday, May 10, 2021 2:40 PM
To: edwardsm@vt.edu
Subject: RE: FOIA

It is my understanding we have delivered to him everything he asked for that is in our possession.

[redacted]

Thanks!

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Monday, May 10, 2021 9:03 AM
To: Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Hi Jamie,

Any luck on finishing this request? Thanks in advance.

Marc

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Thursday, April 22, 2021 2:52 PM
To: edwardsm@vt.edu
Subject: Re: FOIA

Let me send a follow up email. I'll let you know! Thanks!

Sent from Jamie's iPhone

> On Apr 22, 2021, at 12:30 PM, edwardsm@vt.edu wrote:
>
> Hi Jamie,
>
> I never heard back from you. Will we be getting the rest of the laboratory reports and chain of custodies that were missing?
>
> Thanks!
>
> Marc

[redacted] 2/2

VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: FOIA

edwardsm@vt.edu <edwardsm@vt.edu> Thu, May 20, 2021 at 7:08 PM
[redacted] Kory Wait <korydw@vt.edu>

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Thursday, May 20, 2021 7:08 PM
To: 'Vanessa Kemp' <vanessa@cityofmoore.com>
Cc: 'Robert Pistole' <robert.pistole@veolia.com>; 'Brian Miller' <BMiller@cityofmoore.com>; 'Jamie Weaver' <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Attached is the invoice of how you successfully obtained the 2018 records, which we appreciate.

Marc

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Thursday, May 20, 2021 7:01 PM
To: 'Vanessa Kemp' <vanessa@cityofmoore.com>
Cc: 'Robert Pistole' <robert.pistole@veolia.com>; 'Brian Miller' <BMiller@cityofmoore.com>; 'Jamie Weaver' <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Dear Vanessa,

Can you please explain this to me?

We requested specific water analysis records for City of Moore wells from 2018, 2019, 2020.

The year 2018 records were provided to us as requested in the correct format. Thank you.

We then asked for the remaining records from 2019 and 2020. Jamie checked and emailed. She said that I was correct that these were overlooked.

Unfortunately, the records that were sent to us from 2019 and 2020, were not in the format we requested.

[redacted] 1/4

We received the correct format with chain of custody for 2018, so why wasn't it possible to repeat that request for the records requested in 2019 and 2020?

Why are these requested water analysis records not "obtainable," when they were already obtained just fine for the year 2018?

Marc Edwards

From: Vanessa Kemp <vanessa@cityofmoore.com>
Sent: Thursday, May 20, 2021 11:42 AM
To: edwardsm@vt.edu
Cc: Robert Pistole <robert.pistole@veolia.com>; Brian Miller <BMiller@cityofmoore.com>; Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Marc,

I have checked on this and you have received everything that is obtainable in reference to your records request.

Vanessa Kemp, City Clerk

City of Moore

301 N Broadway | Moore, OK 73160 | 405-793-5021

[redacted]

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Wednesday, May 19, 2021 7:34 PM
To: edwardsm@vt.edu; Vanessa Kemp <vanessa@cityofmoore.com>
Subject: Re: FOIA

Hi Marc. If you will remember when I first spoke to you in March, I explained to you that our department had someone out on leave and I was just helping to cover. I will add Vanessa to this email. She is the person you need to speak with.

Thank you!

Sent from Jamie's iPhone

> On May 19, 2021, at 7:04 PM, edwardsm@vt.edu wrote:
>
> Hi Jamie,
>
> It seems like we are coming up on one year waiting for the simple pdfs of the sample analysis and chain of custodies that we requested.
>
> Is there some kind of problem?
>
> You have told me repeatedly that we will get them, and every time I check back it takes weeks.
>
> I paid for them months and months ago.
>
> Best Regards,
>
> Marc

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Monday, May 10, 2021 10:03 AM
To: JWeaver@cityofmoore.com
Subject: RE: FOIA

Hi Jamie,

[redacted] 3/4

Any luck on finishing this request? Thanks in advance.

Marc

From: Jamie Weaver <JWeaver@cityofmoore.com>

[Quoted text hidden]

[Quoted text hidden]



IMG_3574.jpg
240K

[redacted] 4/4

J.4 Summary of correspondence of 2nd FOIA request in Moore, OK

VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: Third FOIA Request

edwardsm@vt.edu <edwardsm@vt.edu> Fri, May 21, 2021 at 3:48 PM
To: Kory Wait <korydw@vt.edu> [redacted]

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Friday, May 21, 2021 3:48 PM
To: 'Vanessa Kemp' <vanessa@cityofmoore.com>; 'Jamie Weaver' <JWeaver@cityofmoore.com>
Cc: 'Robert Pistole' <robert.pistole@veolia.com>; 'Brian Miller' <BMiller@cityofmoore.com>; 'Jamie Weaver' <JWeaver@cityofmoore.com>
Subject: Third FOIA Request

Vanessa,

While I await a response to our second very simple FOIA request, and an answer to my question as to why you think none of these documents are available, I am submitting a new request.

This 3rd FOIA request, asks for documents that have still not been provided in response to my first request. It also seeks new information.

Documents That Exist But Which Are Missing from My First FOIA Request Document Production:

1. I previously requested records that would allow me to calculate the volume of water purchased monthly from Oklahoma City. These records were not supplied and I would like to repeat and rephrase our request with the following specifics:

    a. Drinking Water Monthly Usage Summary that shows: Month, # of Gallons Pumped from wells, # of Gallons Purchased from Oklahoma City, and Total # of Gallons used each month. Please supplies records for the years 2018, 2019, 2020.
    b. I would like documentation that shows the price(s) paid per gallon paid to Oklahoma City for drinking water from 2015 to present.

2. I had previously requested records regarding volume of water drawn from individual city wells from 2015 to present. Such records were not supplied and I would like to repeat and rephrase our request as follows:

    a. Records that show the City of Moore active water wells, by well ID number, for each month from 2016 to present. Please include documents that show the estimated volume of water pumped for each well.

NEW INFORMATION

[redacted] 1/5

3. Documentation of all maintenance performed on Well 023 (Well 24), Well 018 (Well 19), and Well CL037 (Well 37) for 2017 to present.
    a. The documents should include dates and descriptions of procedures such as “cleaning”, “acidizing”, “installation of new pump and motor”, etc.
    b. This request is in reference to cited use of such procedures in the Notice of Violations statements for arsenic sent to Moore residents.

Best Regards

Marc Edwards

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Thursday, May 20, 2021 7:01 PM
To: 'Vanessa Kemp' <vanessa@cityofmoore.com>
Cc: 'Robert Pistole' <robert.pistole@veolia.com>; 'Brian Miller' <BMiller@cityofmoore.com>; 'Jamie Weaver' <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Dear Vanessa,

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Unfortunately, the records that were sent to us from 2019 and 2020, were not in the format we requested.

We received the correct format with chain of custody for 2018, so why wasn’t it possible to repeat that request for the records requested in 2019 and 2020?

Why are these requested water analysis records not “obtainable,” when they were already obtained just fine for the year 2018?

Marc Edwards

[redacted] 2/5

From: Vanessa Kemp <vanessa@cityofmoore.com>
Sent: Thursday, May 20, 2021 11:42 AM
To: edwardsm@vt.edu
Cc: Robert Pistole <robert.pistole@veolia.com>; Brian Miller <BMiller@cityofmoore.com>; Jamie Weaver <JWeaver@cityofmoore.com>
Subject: RE: FOIA

Marc,

I have checked on this and you have received everything that is obtainable in reference to your records request.

Vanessa Kemp, City Clerk

City of Moore

301 N Broadway | Moore, OK 73160 | 405-793-5021

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Wednesday, May 19, 2021 7:34 PM
To: edwardsm@vt.edu; Vanessa Kemp <vanessa@cityofmoore.com>
Subject: Re: FOIA

Hi Marc. If you will remember when I first spoke to you in March, I explained to you that our department had someone out on leave and I was just helping to cover. I will add Vanessa to this email. She is the person you need to speak with.

Thank you!

Sent from Jamie's iPhone

On May 19, 2021, at 7:04 PM, edwardsm@vt.edu wrote:

Hi Jamie,

It seems like we are coming up on one year waiting for the simple pdfs of the sample analysis and chain of custodies that we requested.

Is there some kind of problem?

You have told me repeatedly that we will get them, and every time I check back it takes weeks.

I paid for them months and months ago.

Best Regards,

Marc

From: edwardsm@vt.edu <edwardsm@vt.edu>
Sent: Monday, May 10, 2021 10:03 AM
To: JWeaver@cityofmoore.com
Subject: RE: FOIA

Hi Jamie,

Any luck on finishing this request? Thanks in advance.

Marc

From: Jamie Weaver <JWeaver@cityofmoore.com>
Sent: Thursday, April 22, 2021 2:52 PM
To: edwardsm@vt.edu
Subject: Re: FOIA

Let me send a follow up email. I'll let you know! Thanks!

[redacted] 4/5

Sent from Jamie's iPhone

On Apr 22, 2021, at 12:30 PM, edwardsm@vt.edu wrote:

Hi Jamie,

I never heard back from you. Will we be getting the rest of the laboratory reports and chain of custodies that were missing?

Thanks!

Marc

[redacted] 5/5

VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: OKC water usage invoices-ATTACHED IS DEC 2016 BACK TO JAN 2016 -BATCH #6

edwardsm@vt.edu <edwardsm@vt.edu> Tue, Jun 15, 2021 at 12:01 PM
To: Kory Wait <korydw@vt.edu>[redacted]

From: Brian Miller <BMiller@cityofmoore.com>
Sent: Tuesday, June 15, 2021 11:55 AM
To: edwardsm@vt.edu; Vanessa Kemp <vanessa@cityofmoore.com>; Jamie Weaver <JWeaver@cityofmoore.com>; Robert Pistole <robert.pistole@veolia.com>
Subject: FW: OKC water usage invoices-ATTACHED IS DEC 2016 BACK TO JAN 2016 -BATCH #6

Brian K. Miller

Assistant City Attorney/Risk Manager

City of Moore

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(405) 794-5579

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VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: OKC water usage invoices -ATTACHED IS DEC 2017 BACK TO JAN 2017 - BATCH #5

edwardsm@vt.edu <edwardsm@vt.edu> Tue, Jun 15, 2021 at 12:01 PM
To: Kory Wait <korydw@vt.edu>[redacted]

From: Brian Miller <BMiller@cityofmoore.com>
Sent: Tuesday, June 15, 2021 11:54 AM
To: edwardsm@vt.edu; Vanessa Kemp <vanessa@cityofmoore.com>; Jamie Weaver <JWeaver@cityofmoore.com>; Robert Pistole <robert.pistole@veolia.com>
Subject: FW: OKC water usage invoices -ATTACHED IS DEC 2017 BACK TO JAN 2017 - BATCH #5

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VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: OKC water usage invoices -ATTACHED IS DEC 2018 BACK TO JAN 2018 - BATCH #4

edwardsm@vt.edu <edwardsm@vt.edu> Tue, Jun 15, 2021 at 12:00 PM
To: Kory Wait <korydw@vt.edu>[redacted]

From: Brian Miller <BMiller@cityofmoore.com>
Sent: Tuesday, June 15, 2021 11:53 AM
To: edwardsm@vt.edu; Vanessa Kemp <vanessa@cityofmoore.com>; Jamie Weaver <JWeaver@cityofmoore.com>; Robert Pistole <robert.pistole@veolia.com>
Subject: FW: OKC water usage invoices -ATTACHED IS DEC 2018 BACK TO JAN 2018 - BATCH #4

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VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: OKC water usage invoices - ATTACHED IS DEC 2019 BACK TO JAN 2019

edwardsm@vt.edu <edwardsm@vt.edu>
To: Kory Wait <korydw@vt.edu> [redacted]

From: Brian Miller <BMiller@cityofmoore.com>
Sent: Tuesday, June 15, 2021 11:54 AM
To: edwardsm@vt.edu; Vanessa Kemp <vanessa@cityofmoore.com>; Jamie Weaver <JWeaver@cityofmoore.com>; Robert Pistole <robert.pistole@veolia.com>
Subject: FW: OKC water usage invoices - ATTACHED IS DEC 2019 BACK TO JAN 2019

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From: Pistole, Robert <robert.pistole@veolia.com>
Sent: Thursday, May 27, 2021 8:46 AM
To: Vanessa Kemp <vanessa@cityofmoore.com>; Brian Miller <BMiller@cityofmoore.com>
Subject: Fwd: OKC water usage invoices - ATTACHED IS DEC 2019 BACK TO JAN 2019

---------- Forwarded message ---------
From: **Patti Sandefur** <PSandefur@cityofmoore.com>
Date: Mon, 24 May 2021 at 16:26
Subject: OKC water usage invoices - ATTACHED IS DEC 2019 BACK TO JAN 2019
To: Robert Pistole <robert.pistole@veolia.com>

Patti Sandefur

Accounts Payable

City of Moore

(405) 793-5011

"Disclaimer: This email and any files transmitted with it are intended solely for the use of the individual or entity to which they are addressed. If you have received this email in error ple

--

Robert Pistole

Project Manager

Moore, Oklahoma

(405)793-5087 Office

(405)627-1842 Cellular

(405)793-5094 Fax

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VIRGINIA TECH

Kory Wait <korydw@vt.edu>

FW: OKC water usage invoices - ATTACHED IS DEC 2020 BACK TO JAN 2020 - BATCH #2

edwardsm@vt.edu <edwardsm@vt.edu> Tue, Jun 15, 2021 at 12:00 PM
To: Kory Wait <korydw@vt.edu> [redacted]

From: Brian Miller <BMiller@cityofmoore.com>
Sent: Tuesday, June 15, 2021 11:53 AM
To: edwardsm@vt.edu; Vanessa Kemp <vanessa@cityofmoore.com>; Jamie Weaver <JWeaver@cityofmoore.com>; Robert Pistole <robert.pistole@veolia.com>
Subject: FW: OKC water usage invoices - ATTACHED IS DEC 2020 BACK TO JAN 2020 -BATCH #2

Brian K. Miller

Assistant City Attorney/Risk Manager

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J.5 Supplemental Tables for Chapter 5

Table J.1: Summary of sampling in the Chicago Metropolitan area

Area	**Chicago**	**Berwyn**	**Cicero**	**Robbins**
Total lead connections*	392,614 of 519,711**	3,300 of 3,865	14,872 of 15,175	Unknown of 2,186
Total homes in analysis	194	25	52	68
Homes with concerning* water lead**	**57 (29%)**	**9 (36%)**	**17 (33%)**	**1 (1.5%)**
Highest lead (ppb)	961.3	108	102	18.4
1st draw 90th percentile (Lead and Copper Rule Standard)	8.7	11.5	15.4***	1.5
2nd draw 90th percentile (Lead and Copper Rule Standard)	10.4	12.2	18.7	1.0
3rd draw 90th percentile (Lead and Copper Rule Standard)	11.8	8.9	9.3	1.3
Sampling conducted in:	Jul-Aug 2019	Aug 2018	Aug 2018	Sep 2018

*Illinois EPA: https://www2.illinois.gov/epa/topics/drinking-water/public-water-users/Pages/lead-service-line-information.aspx

**120,760 service lines of unknown material in Chicago

***Violation of Lead and Copper Rule Action Level of 15 ppb