Water Lead Levels of Private Domestic Well-Supplied

Homes in Three Rural Illinois Counties

BY

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THESIS

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JLB

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LIST OF ABBREVIATIONS

ACS	American Community Survey
ADHD	Attention Deficit/Hyperactivity Disorder
ALAD	δ-aminolevulinic acid dehydratase
ANCOVA	Analysis of Covariance
BLL	Blood Lead Levels
BRFSS	Behavioral Risk Factor Surveillance System
CSMR	Chloride to Sulfate Mass Ratio
CWS	Community Water System
DIC	Dissolved Inorganic Carbon
EPA	United States Environmental Protection Agency
EPM	Equivalents Per Million
FFQ	Food Frequency Questionnaire
GSH	Glutathione
HDPE	High Density Polyethylene
HPT	Hypothalamus-pituitary-testicular
IARC	International Agency for Research on Cancer
IDPH	Illinois Department of Public Health
IRB	Institutional Review Board
ISWS	Illinois State Water Survey
LHD	Local Health Department
LSI	Larson Skold Index
MAC	Maximum Acceptable Concentration
ND	Not Detected
NHANES	National Health and Nutritional Examination Survey
NPV	Negative Predictive Value
PDW	Private Domestic Well
PKC	Protein Kinase C

LIST OF ABBREVIATIONS (continued)

POE   Point of Entry

POU   Point of Use

PPV   Positive Predictive Value

QA/QC   Quality Assurance/Quality Control

ROC   Reactive Oxygen Species

SDWA   Surface Drinking Water Act

SMCL   Secondary Maximum Contaminant Level

TDS   Total Dissolved Solids

TEL   Tetraethyl Lead

WLL   Water Lead Levels

SUMMARY

Background: Studies in Washington DC, Montreal, and most recently in Flint, MI have been a wake-up call for cities and researchers to focus on the impacts of the corrosion of old service lines, premise piping, and solder on water lead levels, and their contributions to population and community-wide lead intake. However, there has been less focus on the lead levels resulting from groundwater corrosion of private domestic wells.

Private well baseline surveillance is minimal, since wells are not subject to Federal regulatory monitoring requirements attached to laws and statutes like the Safe Drinking Water Act (SDWA). There are some requirements at the state and local level for private well testing, including Illinois, but these do not include lead testing. State and community-level studies in Virginia, Pennsylvania, Wisconsin, and New York have uncovered elevated water lead levels, but only one of these studies appropriately characterize corrosion mechanisms and their relationship to elevated water lead levels.

Research Question: We sought to evaluate corrosivity and housing age (as a proxy for aging plumbing and well components) as possible predictors of total water lead in private well-supplied homes in rural Illinois. We expected that more corrosive water, interacting with older homes and wells, would produce higher lead levels at the tap. We hoped to ascertain if these predictors could be used as a decision rule for a local health department to prioritize homes for testing.

Methods: Using a cross-sectional design, we teamed up with local health departments (LHDs) of three largely rural Illinois counties (Kane, Peoria, and Jackson). Following a protocol approved by the UIC Institutional Review Board (IRB), LHD staff assisted with the enrollment of a convenience sample of county residents who rely on domestic wells for their drinking water. Participants collected water samples, which were analyzed for lead and corrosivity-related parameters (chloride, sulfate, and alkalinity), and indicated the year of construction for their home. We conducted univariate and bivariate analyses to characterize the relationships of water lead levels,

corrosivity (measured with the Larson Skold Index (LSI) and the Chloride to Sulfate Mass Ratio (CSMR)), and age of housing. We stratified specifically by homes built before/in 1986. Additional analyses included logistic regression modelling to evaluate the presence of an interaction effect, and checking the diagnostic utility of our final decision rule.

Results: About 47% (n=97) of our first draw (stagnant) samples had detectable levels of lead, with only 3.1% exceeding the US Environmental Protection Agency (EPA) regulatory levels of 15 µg/L. When evaluating corrosivity indices for use, we found that the Larson Skold Index did not vary significantly by county, while the CSMR was significantly higher in Kane County than Jackson County (p<0.0001). There was no association between corrosivity and housing. We found significant differences in the median water lead levels of homes built before and after 1986 (before: 1.11, after: 0.54, p=0.01). There were statistically significant associations between LSI empirical cutoffs and lead detection and exceedance of 1.57 µg/L (67th percentile).   Similar significant associations were found using CSMR thresholds, but that relationship remains poorly understood. Logistic regression modelling revealed stratum specific significant effects of lead detection (OR: 4.13, 95% CI: 1.02, 16.68) in the presence of pre-1986 homes and Larson Skold Index values exceeding 0.59 (the 75th percentile). The modelled decision rule had low sensitivity (31.0%).

Discussion and Public Health Implications: This pilot study may serve as the basis for a broader analysis of Illinois private domestic wells. Limitations to this study include low generalizability, reliance on participant sampling and recall, corrosivity metrics limitations, and lack of direct plumbing/well inspections. Our results are consistent with other similar studies. These findings may encourage states (such as but not limited to Illinois) to consider expanded private well testing regulations and pre-existing subsidized testing programs. In particular, such surveillance may help alleviate inequities in health between rural and urban communities in Illinois.

I.   AN OVERVIEW OF GENERAL LEAD EXPOSURE

A. Toxicology and Health Effects of Lead

Lead is a toxicant known for established associations and associations currently under study with numerous neurological, cardiovascular, nephrotoxic endpoints. According to National Health and Nutritional Examination Survey (NHANES) data, measured blood lead levels have been on the decline since the 1970s, concurrent with bans on lead paint and lead in gasoline. Nonetheless, there is no safe level of lead in blood, and there are still inequities in lead exposures in the population, particularly by income and by race/ethnicity (Moody, Darden, & Pigozzi, 2016).

Lead has multiple mechanisms of toxicity. Lead is particularly effective at inhibiting hemebiosynthesis, and hemoglobin production as a result. One of the enzymes susceptible to lead is δ-aminolevulinic acid dehydratase (ALAD), which generates a heme precursor. The enzyme ALAD can be inhibited over a large range of blood lead levels starting at <10 µg/Dl (Agency for Toxic Substances and Disease Registry, 2007). This impact is magnified through inhibition of glutathione (GSH), an enzyme that stabilizes reactive oxygen species. The sum total of both GSH and ALAD inhibition, with the destabilization of cellular membranes via lipid peroxidation, results in hemolytic anemia and oxidative stress-induced cell death. One of lead's prominent neurotoxic mechanisms of action is through mimicking calcium in neuron cell uptake and displacing it from its binding sites. This allows lead to pass through the blood brain barrier and subsequently hurts mechanisms managing the maintenance of synapses, neuronal development, and neuron and glia interactions (Kim et al., 2015; Vorvolakos, Arseniou, & Samakouri, 2016). Increased Protein Kinase C (PKC), which is regulated by Ca^{2+}, can impair brain microvascular formation and function, as well as disrupt pre-frontal cortical regulatory activities. It also impacts the various neurotransmission systems (glutamatergic, cholinergic, and dopaminergic), most specifically the glutamatergic. Interestingly, lead seems to have a biphasic dose-response in this regard, as suppression of glutamatergic system seems to occur predominantly at lower or at high doses.

The increased amount of oxidative stress also contributes to increased neuron vulnerability (Vorvolakos et al., 2016).

Lead also has carcinogenic and mutagenic mechanisms of interest, but these are currently not fully understood. Toxicology studies on rodents found accelerated kidney tumor growth similar to the growth initiated by other carcinogens as well. Additionally, higher incidence of kidney and liver tumors were seen after being fed both lead subacetate and 2-acetylaminofluorene when compared to rodents with a diet of either lead subacetate or 2-acetylaminofluorene (IARC & IARC Working Group on the Evaluation of Carcinogenic Risks to Humans, 2006). Damage to DNA and fragmentation have been observed in *in vivo* studies, but *in vitro studies* yielded conflicting results. Based on the current state of research, lead's carcinogenic mechanism in humans appears to be a product of multiple indirect mechanisms related to inhibited DNA synthesis and repair and interacts with tumor-suppressor proteins and DNA-binding proteins (National Toxicology Program, 2016).

1. **Evaluation Standards and Biomarkers**

The primary way of measuring lead exposure is through evaluating blood lead levels (BLL). This metric is an evaluation of both recent and longer term exposure, using the lead content of whole blood from an individual. At present, no safe BLL has been discovered for children or adults. The Centers for Disease Control (CDC) currently has a reference level for children of 5 µg/dL, reflective of the upper 97.5th percentile of NHANES. The National Institute of Occupational Safety and Health (NIOSH) has established the same 5 µg/dL BLL as the adult reference level (Raymond & Brown, 2017).

The other biomarkers are used to evaluate lead exposure with limited clinical relevance. One such example is the evaluation of erythrocyte protoporphyrin and zinc protoporphyrin. These two compounds that are formed when lead inhibits ferrochelatase from adding iron into protoporphyrin IX to create heme. This diagnostic tool is only useful when adult BLLs are greater than 30 µg/dL, or when

children BLLs are greater than 15 µg/dL. Additionally, heme reductions are not necessarily a unique metric for lead poisoning. The other evaluation standard is by evaluating the lead in bone. It is measured through the use of x-ray fluorescence of the tibia (Somervaille et al., 1988). This is considered a longer term, cumulative exposure metric, as lead is partitioned into the trabecular bone, which can readily be assimilated into the blood, and the more stable cortical bone. Thus, elevated blood lead levels may be reflective of bone-partitioned lead through this interchange. However, this is considered more of a research metric than a practical clinical measure.

2. **Acute Effects of Lead**

Many of the clearly observable acute effects are neurologic/neurobehavioral, or cerebrovascular in nature. Male workers experiencing exposures 40-60 µg/dL had impaired abilities in verbal concept formation, visual/motor skill performance, memory, and mood, but did not present any particular peripheral nervous system impacts. At high levels (100-120 µg/dL for adults, 80-100 µg/dL for children) often result in encephalopathy, and severe delirium, , often leading to coma and death (Vorvolakos et al., 2016). Other impacts include more mobility-based impacts such as postural balance and paresthesia.

3. **Chronic Health Effects in Adults**

i. **Cardiovascular Effects**

Animal model studies have identified a plausible mechanism of how lead could cause hypertension through neurohumoral regulation of vascular resistance, heart rate, and overall cardiac output (Carmignani et al., 2000; Khalil-Manesh et al., 1993; Ni, Hou, Barton, & Vaziri, 2004; Vaziri & Sica, 2004). Recent prospective studies using NHANES-III data have found significant positive associations between diastolic blood pressure and exposure levels of BLLs (0-2 µg/dL, 2-5 µg/dL, 5-10 µg/dL, and 10+ µg/dL) (Obeng-Gyasi et al., 2018), as well as significant positive associations between increases in BLL

from 1.0 μg/dL to 6.7 μg/dL (the 10th-90th percentiles of BLL in NHANES) and increases cardiovascular ischemic heart disease mortality (Lanphear, Rauch, Auinger, Allen, & Hornung, 2018).

ii.   Nephrotoxic Effects

Much of the nephrotoxic impacts of lead can be related to oxidative stress from reactive oxygen species (ROS), and as well as causing lipid oxidation and DNA fragmentation. In particular, lead seems to have strong mitochondrial impacts affecting both membrane behavior and overall osmotic balance of renal cells, causing membrane potential change through replacing Ca^{2+}, apoptosis initiation, and rupture of the outer membrane (Orr, Bridges, Orr, & Bridges, 2017).

A suite of health effects has been observed in a dose response relationship with BLLs. BLLs <20 μg/dL have been associated mostly with impacts on glomerular filtration, while those >30 μg/dL have been associated with enzymuria and proteinuria (increased enzyme and protein content of the urine, respectively). Above 50 μg/dL, major functional and pathological changes start becoming increasingly evident. When adjusting for age, decreased glomerular filtration has repeatedly been surveyed in populations with BLLs <20 μg/dL. Additional studies have found these same associations lower than 10 μg/dL (Agency for Toxic Substances and Disease Registry, 2007). However, it is important to note that hypertension may be a cause or consequence of lead, as decrements in glomerular filtration can increase blood pressure.

iii.   Infertility

Lead is able to adversely impact what is known as the "hypothalamus-pituitary-testicular (HPT) axis", the sequential interaction of the hypothalamus and the pituitary gland that ultimately affect the behavior of the gonadal system of men. Lead impairs signaling pathways and the receptors of the pituitary and the gonadal systems, respectively, preventing induction of spermatogenesis processes. Additionally, as ascertained through animal studies, lead may more directly affect overall

spermatogenesis, resulting in immature sperm cells, lowered sperm cell function, and low sperm count. In particular, generation of ROSs can specifically alter sperm viability, motility and other functions that can reduce fertilization rates. In epidemiology studies, BLLs of $\geq$ 40 μg/dL have been found to impact male fecundity, quantified by the time to pregnancy of their respective partners. The HPT-axis is relatively vulnerable, and may be irreversibly impacted by lower dose chronic lead exposures than high doses (Gandhi et al., 2017).

iv.   Carcinogenicity

Inorganic lead has put in Group 2A (probably carcinogenic) by the International Agency for Research on Cancer (IARC) based on animal testing and limited data from human subjects. Cancers linked to inorganic lead exposure include stomach, bladder, and lung cancers. Organic lead, by contrast, is in Group 3, meaning that there is not sufficient evidence to evaluate its impacts (IARC & IARC Working Group on the Evaluation of Carcinogenic Risks to Humans, 2006; National Toxicology Program, 2016) .

v.   Neurological

Most neurological effects resulting from lead exposures can be traced back to childhood exposures (discussed in the following sections). Case studies have demonstrated that reducing elevated lead body burden has resulted in mitigating depression symptoms (Sohler, Kruesi, & Pfeiffer, 1977).

4. Chronic Health Effects of Children

While many of the effects of children and adults are similar, child exposures are greater because of their increased contact with lead sources and higher uptake rates in relation to their body weight. Ingestion and inhalation are the main pathways to exposure of lead, and children are more likely to ingest lead-containing soil, dust, and paint chips. Children may experience a greater internal dose relative to their size. Finally, many child exposures may cause irreversible developmental impacts that may be precursors to additional adult conditions.

i. Neurological Consequence in Adulthood of Childhood Lead Exposure

The most widely studied impacts from lead result from its neurotoxic impacts. Chronic lead exposure is associated with various neurobehavioral impacts, such as distractibility, impulsivity, and shortened attention span in children. While the magnitude of association is still under investigation, Attention Deficit/Hyperactivity Disorder (ADHD) has been reliably associated with 10μg/dL and, to a certain extent, BLL under 5 μg/dL (Agency for Toxic Substances and Disease Registry, 2007; Vorvolakos et al., 2016). Results from cohort studies for 6 months-5 year olds indicate that increase in 10 μg/dL for children can results in 4.6 point decreases in IQ measured by the Stanford-Binet Intelligence Scale. For children with BLLs below 10 μg/dL , IQ declined by 7.4 points with every 1 μg/dL increase in BLL (Canfield et al., 2003).

A recent cohort study of children found that children with lifetime average blood concentrations between 5-9.9 μg/dL scored 4.9 points lower on the Wechsler Preschool and Primary Scale of Intelligence–Revised, and that this inverse association between blood lead and "Full-Scale IQ" was observed at peak BLLs as low as 2.1 μg/dL (Jusko et al., 2008). Cross-sectional studies using NHANES data, among others, have found associations between elevated BLLs and having major depressive disorder and panic disorder (Sohler et al., 1977; Vorvolakos et al., 2016).

Many of these neurological impacts may be developmental in nature, with exposures carrying over from maternal exposures, or not manifesting until later in development. A prospective study (Opler et al., 2004) found that increased levels of δ-aminolevulinic acid (ALAD substrate) in maternal serum was associated with perinatal lead exposure as low as 15 ug/dL and the later development of schizophrenia. Higher mean childhood concentrations have also been associated with reduced adult grey matter in specific sections like the anterior cingulate cortex, which may be related to cognitive and behavioral issues.

B. Water as a Source of Lead

    1. Historical Environmental Lead Sources

Historically, the most common media of residential environmental exposure which people, specifically children, come into contact with lead are from lead dust, soil, and paint. One of the main sources of lead came from the use of tetraethyl lead (TEL) in gasoline, which was introduced in the 1920s as an anti-knocking agent. While its health impacts were questioned and scrutinized at first, TEL became widespread worldwide (Hernberg, 2000). TEL contributed to airborne lead exposure through tailpipe emissions, and contributed to soil and dust pollution through depositional and windblown processes. The other source, through paint, was widely used because of its durability. Childhood lead poisoning and lead poisoning related symptoms from paint were first prominently documented by Turner (1897) and Gibson (1908), who both observed that afflicted child patients were ingesting lead paint off of their fingers. Turner, in particular, called for primary prevention and elimination of lead paint. These two findings eventually spurred the International Labor Organization (then the International Labor Convention) to put forth the White Lead Convention in 1921. Multiple European countries banned the use of white lead paint at this time, but more pertinently, the United States did not do so until 1978 (Hernberg, 2000).

Individuals today can still be exposed directly to lead paint through older housing stock, or through dust that is deposited off of lead paint. While some children are directly exposed to peeling paint, its major pathway that contributes to childhood lead poisoning is through its influence on indoor dust, as confirmed in Lanphear et al. (1998). At the federal level, if a home was built before 1978, property owners with lead-painted properties are required to inform prospective buyers and tenants about the risk in accordance with the Residential Lead-Based Paint Reduction Act (enacted in 1992), but this law does not require the landlord to remove the lead paint itself (United States Housing and Urban Development, 2019). However, there are some additional Federal laws and regulations in place that do

have some enforcement of lead paint removal and protection. The Environmental Protection Agency's

(EPA) Renovation, Repair, and Painting rule (enacted 2008, enforced in 2010), requiring contractors to

be certified and use lead safe practices that reduce potential contamination from disturbed lead paint

(United States Environmental Protection Agency, 2013). Additionally, any federally owned or subsidized

housing is subject to the Lead Safe Housing Rule (enacted in 2000), with specific lead hazard mitigation

and control requirements, as well as mandating notification of hazard control and evaluation (United

States Housing and Urban Development, 2019). There are further regulations regarding lead paint

mitigation at the state and municipal level, as well as programs to help with lead cost and remediation.

In the state of Illinois, landlords are required to test houses with elevated BLL children for lead paint,

and must mitigate accordingly and inform all tenants of the building of mitigation activities. In the City

of Chicago as an example, the Department of Public Health has some grant funding to assist low-income

home owners and landlords with shouldering the cost of mitigating lead hazards (Lead Safe Illinois,

2019). In both Chicago and Illinois, there are no restrictions of selling real estate with lead paint.

2. Major Water Lead Studies

Drinking water as a source of lead received particular scrutiny when widespread elevated lead

levels were evaluated in the crisis of lead in drinking water of Washington, DC. The "DC Water Crisis"

began in 2001 as a consequence of changing the drinking water disinfection method from chlorine to

chloramine. This was done by the Washington Area Sewer and Water Authority (now known as DC

Water) in order to comply with new EPA regulations in 2001. However, this was done without the

addition of corrosion control, resulting in the dissolution of protective scale that had accumulated within

old lead pipes and dissolving the pipe walls. A later, similar crisis occurred in Flint, MI, when their water

source was switched from Lake Huron and the Detroit River to the Flint River in 2014. This switch,

similarly, increased corrosivity of the water and was carried out without corrosion control, causing lead

to leach from pipes into drinking water.

Regarding Montreal, the third major site of municipal drinking water lead research, Health Canada set a health effects-informed Maximum Acceptable Concentration (MAC) of 10 µg/L in 1992 and requires utilities to conduct active monitoring at residential locations annually. Sampling conducted in 2005 found moderate lead concentrations in Montréal households with lead service lines. Authorities at the Montréal Public Health Department used these data to estimate potential BLL increases, finding that the increases were under the notification level of 10 µg/L at the time (Levallois et al., 2014).

Multiple studies have emerged from these three locations analyzing the impacts of water lead levels (WLL) on blood lead (Edwards, Triantafyllidou, & Best, 2009; Gómez et al., 2019; Hanna-Attisha, LaChance, Sadler, & Champney Schnepp, 2016; Levallois et al., 2014; Ngueta, Abdous, Tardif, St-Laurent, & Levallois, 2016). The results of these studies are summarized as follows in TABLE I.

These studies, to varying ability and success, address the hypothesis that increased water lead results in elevated BLLs. Edwards, Triantafyllidou, and Best (2009), in their Washington DC study, demonstrated that there is a dose-response like relationship between the 90th percentile water lead levels and percentages of children aged 1.3 years and younger with elevated blood lead levels (higher than 10 µg/dL). Neighborhoods were evaluated by risk using the percent of homes with lead pipe multiplied by the percent of homes with first draw (stagnant overnight or for a period of hours) WLLs exceeding 100ppb, and were ranked as high, moderate, and low risk. Significant correlations between WLLs and elevated BLLs were observed in high (2.4 times higher) and moderate risk (1.12 times higher) neighborhoods, with the slope of the correlation increasing in higher risk neighborhoods. In Montreal, Levallois et al (2014) observed a significant association (OR: 4.7, 95% CI: 1.3-7.8) between the upper tertile of measured WLL (3.3 µg/L) with the 75th percentile of BLL (1.78 µg/dL), after adjusting for measured paint and dust lead exposures. This study is complemented by work done by Ngueta et al (2016), which modelled how cumulative water exposure may increase BLL with the same data. Using linear regressions to compare BLL to cumulative water exposure, this study found that BLLs had

significant associations with cumulative exposures starting at 0.7 µg Pb/kg body weight. In Flint, Hanna-

Attisha et al (2016) tried to make a similar point, but the researchers did not report the water lead levels

at individual homes. Rather, they used coarser classifications with secondarily collected data on percent

of homes exceeding 15 ppb, using these to then demarcate "high WLL" and "low WLL" areas. From

there, the researchers compared the percent of elevated blood lead levels in the "higher risk" areas to

the "lower risk" ones pre and post the water source shift. They found statistically significant increases in

incidence of elevated BLLs higher than 5 µg/dL overall (+2.5%) and in "high WLL" neighborhoods

(+6.6%).

TABLE I-SUMMARY OF MAJOR LEAD IN COMMUNITY WATER SYSTEM STUDIES

Study	Location	Period of Study	Type	Age Range	Water Lead Levels	Blood Lead Metric
Edwards, Triantafyllidou, and Best, 2009	Washington DC	1999-2007	Retrospective, Ecologic	$\leq$ 1.3 years	90th percentile levels by zip code	Percent of elevated BLLs above 10 µg/dL from individual-level health record data
Hanna-Attisha, LaChance, Sadler, Champney Schnepp, 2016	Flint, MI	April 2014-Oct. 2015	Multiple sequential cross-sectional	$\leq$ 5 years	Wards classified by percent homes exceeding 15 ppb	Percent of elevated BLLs above 5 µg/dL from individual-level health record data
Gómez et al, 2019	Flint, MI	2006-2016	Retrospective analysis of BLL data	$\leq$5 years	Not Provided	Geometric mean levels and percent exceedances during three 18-month periods
Levallois et al, 2014 Ngueta et al, 2016	Montreal, QC	2009-2010	Cross-sectional	1-5 years	Sampled from each participants' home in 30 minute intervals	Directly sampled from participants

These changes stand in contrast to observations of Gomez et al (2019) who reported that BLLs in Flint actually declined over the 2006-2016 period overall and within the "high WLL" wards before, during and after the water source shift. WLLs in Flint were more directly evaluated and presented in Pieper, Martin, et al (2018).

These studies are limited in their ability to relate changes in water lead levels to changes in blood lead. First, neither Hanna-Attisha, et al. (2016) nor Gomez, et al. (2019) evaluated WLLs at the household level. This is particularly glaring for Hanna-Attisha, et al. (2016), where the researchers used a pre-existing sample exceedance map depicting the percent of homes by ward exceeding 15 ppb. This obscures the connection between elevated water lead levels and the Flint River switch because of a lack of granularity. Similarly, while Edwards et al. (2009) do have granular water lead level data, they do not explicitly connect residential concentrations with elevations in blood lead levels, which does not necessarily speak to biological plausibility. Neither Hanna-Attisha, et al. (2016) nor Edwards, et al (2009) display the overall distribution of blood lead levels, with both studies merely looking just at the percentage of elevated blood lead levels. Gomez, et al. (2019) is a direct response to Hanna-Attisha, et al. (2016), and cites the lack of clearly defined blood lead level distribution, showing how this distribution has changed between 2006 to 2016 and has actually declined globally. Finally, Edwards, et al. (2009) and Hanna-Attisha, et al. (2016) are ecological studies that do not evaluate associations between household-level WLLs and individual BLLs. Levallois, et al. (2014) and Ngueta et al. (2016) both are cross sectional studies that explicitly look at WLLs and BLLs at the individual level for children for a defined period of time. They are the most useful study in terms of directly understanding the relationship between WLL and BLL, particularly in the context of lower concentrations.

3. Corrosivity: A Determinant of Water Lead in Plumbing Systems

Unlike other forms of pollution, lead usually does not enter drinking water sources because of a sudden release from a point source, but is because of an interaction between the water itself and the

pipes, fixtures, and solder through which it is comes into contact. Specifically, the mechanism of water

lead levels leaching into drinking water is a product of water corrosivity. Information surrounding water

corrosivity in community water systems has been extensively documented (Cantor, 2011; Hill & Giani,

2011). The mechanism behind corrosion at its core is an electrochemical reaction; different parts or

walls of the piping are the anodic and cathodic sites and flowing water is the medium that enables

electron transfer. This redox reaction results in metal pipe walls and components that can subsequently

interact with different aqueous species, yielding products that may be soluble. Corrosion, therefore, the

dissolution of these resulting soluble products. Corrosion can happen uniformly, where microscopic

anodic and cathodic sites move around the pipe and result in a consistent loss of metal all around, or

non-uniform, where the anode and cathode are relatively fixed and corrosion is more localized.

Methods to prevent corrosion are either controls on pH and other parameters that promote redox

reactions, or the generation of protective insoluble precipitates known as chemical scales.

Corrosive water, therefore, is determined by several interdependent chemical parameters. The

most important parameter, which seems to influence all other parameters, is pH. In municipal systems

particularly, higher pH values reduce the dissolution of metals from the pipe wall, controlling for other

parameters and aqueous species. pH also has significant impacts on total dissolved solids, alkalinity,

total dissolved inorganic carbon (DIC), and more. Alkalinity, a measure of total carbonate, bicarbonate,

and hydroxide, describes the ability of water to neutralize acid and resist changes in pH. Another

measure, total dissolved solids (TDS), indicates the concentration of ionic species and is used as a

measure for conductivity. Higher TDS values can exacerbate already occurring corrosive reactions.

Lower TDS values, particularly for lead, can oxidize metals while trying to reach electroneutrality, as well

as dissolve precipitated films and scales. Dissolved inorganic carbon, which is similar but not identical to

alkalinity, measures levels of carbonate, bicarbonate, and carbon dioxide. It measures constituents that

contribute to alkalinity and has a complex impact on corrosivity.

Other factors that may also be consequential in terms of corrosion that liberates lead include factors that influence galvanic corrosion. Galvanic corrosion occurs when two different metals with different electrical potentials are connected to each other in the presence of an electrolyte. This happens in instances where lead solder joints are connected to copper pipes, or when there is a partial lead pipe replacement. In the former case, the lead components, exposed to a lower pH with lower alkalinity, forms an anodic site that donates electrons favorably to copper. Oxidized lead in these contexts forms one of two complexes: lead (II) sulfate ($PbSO_4$), which is insoluble, and lead (II) chloride ($PbCl_2$), which is soluble. Thus, levels of sulfate and chloride may act on components of the water system, making them function as determinants of corrosion as well as sources of lead.

4. Sources and Forms of Lead in Water

The main sources of water lead in community systems have been mostly attributed to lead service lines, which are pipes that connect a given location to major water distribution mains, and premise (home) plumbing. There are an estimated 6.1 million lead service lines (full and partial) in the United States, with about 11,200 community water systems containing meaningful amounts of lead service lines (Cornwell, Brown, & Via, 2016). Service lines have been estimated to contributed on average 50-75% of all water lead measured at the tap, while premise plumbing has been estimated to contribute 20-35% of all lead on average (Maynard et al., 2008). Lead service lines may contribute lead through the dissolution of pre-existing scales as well as direct oxidation of pipe walls, disruption of scale formation processes and through physical processes that may liberate particulates. The presence of lead service lines may contribute additionally mobilized lead to a premise plumbing system, becoming trapped by scales that form as part of it that eventually come out when conditions change (Maynard et al., 2008). Lead service line contributions are also altered by the length and diameter of the piping in question. Premise plumbing, by contrast, consists of service line-like components, interior piping, various faucets and fittings, and any of the solder used to join any of those components. Many of these

components are made up of alloys and mixtures, such as brass and galvanized pipe, which may contain variable amounts of lead. For example, until the passage of the Reduction of Lead in Drinking Water Act in 2014, brass could legally contain up to 8% lead, in addition to other metals like copper and zinc (Minnesota Department of Health, n.d.).

Waterborne lead comes in two forms: dissolved and particulate. Dissolved lead typically accumulates in water as a product of stagnation time, and is often linked to the presence of lead services lines and lead containing- components of premise plumbing. By contrast, particulate lead comes from chipping lead solder and aging pipes and is more related to physical disturbance and abrasion, which makes it difficult to predict rates of accumulation and release. Therefore, particulate lead may be more related to flow rate, hydraulic regime, and particle characteristics (Deshommes, Laroche, Nour, Cartier, & Prévost, 2010).

5. Mitigation Strategies for Community Water Systems

Levallois et al (2018) discuss the range of mitigation options available for community water systems. Mitigation strategies for helping reduce exposure to lead revolve around either removing lead sources, or minimizing the impacts of leaching. To prevent the release of lead into water, municipalities can physically remove the sources of contamination through partial or full service line replacement, and replacing lead-containing plumbing and fixtures. While both of these options do remove the source of the lead contamination, they can be incredibly expensive. Replacing all lead service lines, with no cost to homeowners, cost the city of Spokane, WA, $3 million, and cost Lansing, MI $44.5 million (Environmental Defense Fund, 2019). Additionally, service line replacements in the short term in the short term they may cause spikes in lead by remobilizing particulates, and partial line replacements themselves create the conditions for galvanic corrosion to occur. The other method of preventing the release of lead into water is through the use of corrosion control agents, such as orthophosphates, to reduce corrosive conditions and impacts on leaded pipe and components. However, before

implementing corrosion control, one needs to conduct water chemistry testing beforehand, and requires monitoring to ensure conditions are stable.

Another class of mitigation strategy includes simply reducing the impacts of lead leaching on water for consumption. Methods that follow this track include only using cold water taps or using flushing techniques before using water. Both of these techniques are fairly easy to use, but do not address the underlying contamination source, and these type of behavioral interventions are hard to enforce consistently. In the case of flushing, building specific conditions need to be taken into account.

A third class of strategy that is a merger of both is the use of filtration technologies, whether at the point of use (POU), i.e., the outlet at which water is consumed, or at point of entry (POE), i.e., where water enters an individual household or building. These are also rather easy to implement and quite effective, but they require timely maintenance and replacement, meaning that their efficacy needs to be consistently monitored over time.

II.   PRIVATE DOMESTIC WELL WATER AS A SOURCE OF LEAD

A. <u>Introduction</u>

There is currently no federal level regulation of private domestic wells. They are not within the purview of EPA, and EPA does not have any promulgated standards for levels of given contaminants. The CDC does have a program to help state health departments reduce harmful well exposures via the Safe Water for Community Health, which provides technical guidance in developing more robust policy and practice. However, this program is optional and would require states to carry out the recommendations themselves.

There are some state regulations of domestic private wells, but they vary from state to state. As of 2018, there are currently 18 states that have some form of regulation on the books (Schneider, 2019). About 17 states (including Illinois) require at a minimum testing after well construction, 8 require testing after well repair, and 3 require testing before real estate transactions. These regulations, however, mostly concern regulating well testing for a limited scope of contaminants by the owner. In the case of Illinois, county health departments are required to sample new wells for coliform bacteria and give well owners those test results after well construction. The Illinois Department of Public Health (IDPH) is then charged with informing owners of the significance of the results and giving recommendations, which owners must comply with or will face enforcement action.   Many counties also require nitrate testing and testing of other species (A. Schneider, personal communication).

In the United States, it is estimated that around 13.5 million households, or around 42.5 million people (13% of the population) use private wells from for drinking water (Dieter & Maupin, 2017; US Census Bureau, 2017). It is estimated that about 20% of these wells have contaminant levels above at least one current EPA drinking water standards for community water systems (Farquhar, 2018). The demographics of well owners nationwide are detailed in TABLE *II*. The majority of well owners (61.69%)

have at most a high school diploma, and about 14% of well owners were living below the 2017 poverty

threshold for an average family of three ($19,515). Thus, well ownership is a significant problem for

rural populations, who may already be disadvantaged socioeconomically.

TABLE II- DEMOGRAPHICS OF WELL OWNERS BASED ON US AMERICAN HOUSING SURVEY

Well Owner Demographics	N	% of Well Owners
Number of Well Owners	13530	
Race		
White	12640	93.42%
Black	501	3.70%
Hispanic	577	4.26%
Asian/Pacific Islander	94	0.69%
Other	290	2.14%
Income		
<$10,000,	856	6.33%
$10,000-20,000,	1031	7.62%
$20,000-30,000,	1262	9.33%
$30,000-39,999	1291	9.54%
$40,000-50,000,	1211	8.95%
$50,000-60,000,	1094	8.09%
$60,000-80,000,	1846	13.64%
$80,000-100,000,	1483	10.96%
$100,000-120,000,	1084	8.01%
$120,000+,	2366	17.49%
Education		
Less than 9th grade	365	2.70%
Incomplete High School	966	7.14%
High School Diploma	7015	51.85%
Associate Degree	1447	10.69%
Bachelors	2306	17.04%
Graduate/Professional Degree	1426	10.54%

B. Illinois Domestic Well Use and Presence

According to 2015 results from the Illinois Behavioral Risk Factor Surveillance System (BRFSS) (TABLE *III*), about 700,000 adults (estimated 8.60%) use a private well as their main drinking source. The majority of well owners are white, and 40% of them make earn less than $50,000 a year. Educationally, about 41.4% of them do not have beyond a high school diploma. Additionally, the majority of them own the homes in which they live. Thus, while many of them may have the ability in terms of property rights to make adjustments to their well or plumbing to mitigate lead exposure, many of them may not know what exactly to do or have the means to make this a reality.

C. Types of Wells

There are four major types of private drinking water wells: drilled, driven, bored, and dug. Drilled wells are typically the deepest, and can reach depths in excess of 1000 feet. They require the largest amount of effort to install. One method of installation, the "cable tool method", is where a heavy chisel bit is raised and dropped by a steel chain to break through the rock and sediment below before pounding the casing into the rock. The other, more common method is the rotary method, where a long hollow drill tip is filled with a clay slurry to force out debris as drilling commences. They have a lower risk of contamination because of their depth and the continuous casing. Additionally, they are better at handling high water demands.

Driven wells, also known as drillpoint or sandpoint wells, have a small diameter. These are constructed in sand and gravel areas. They consist of tightly connected steel pipes that have a pointed screen at the bottom. One drives the thin well casing pipe into the ground until the point is under the water table, and the screen is able to allow in water and keep sand and gravel out. They are more susceptible to contamination because they are found in areas with sandy soils that are more permeable with a higher water table, meaning that pollution can more easily be conducted through the aquifer.

TABLE III- DEMOGRAPHICS OF ILLINOIS ADULTS USING PRIVATE WELLS AS HOME WATER SOURCE

Demographics[1]	Composition of Well Owners	
Age	**N**	**%**
(n=693,476)		
18-24	78,932	11.38%
25-44	144,625	20.86%
45-64	299,309	43.16%
65+	170,610	24.60%
Race		
(n=647,693)		
White	610,822	94.31%
Black/African American	21,124	3.26%
Other	15,747	2.43%
Hispanic/Latin Descent		
(n=693,915)		
Yes	39,951	5.76%
No	653,964	94.24%
Gender		
(n=697,467)		
Male	341,941	49.03%
Female	355,526	50.97%
Income		
(n=647,976)		
< $15,000	46,106	7.12%
$15-35,000	90,919	14.03%
$35-50,000	119,146	18.39%
> $50,000	391,805	60.47%
Education Status		
(n=697,476)		
<High school graduate	42,813	6.14%
High school graduate	245,783	35.24%
Some college	237,445	34.04%
College graduate	171,426	24.58%
Home Ownership		
(n=693,122)		
Own	582,622	84.06%
Rent	67,678	9.76%
Other arrangement	42,822	6.18%

[1]Source: 2015 Illinois Behavioral Risk Factor Surveillance System, data acquired from the Illinois Department of Public Health

Bored wells are constructed in lower, shallower aquifers, and are typically 30-100 ft. deep. They

are used in low yielding aquifers, and as such they have a large diameter to increase the volume of a

storage reservoir for high demand periods. These are constructed by using an earth augur to bore a

hole in the ground, which is then lined or cased in concrete curbing, stone-derived material, or a well

casing. Similarly, dug wells are constructed by digging a hole town to the water table, and also have a

wide diameter. The walls of the dug wells are often lined with rock, brick, wood, pipe, and other related

materials. These are similarly shallow wells. Both bored and dug wells are more vulnerable to

contamination from runoff and other inputs by virtue of being shallower.

D. Current State of Knowledge of Water Lead Levels in Private Wells

Little is known about the distribution of WLLs in the tap water of US homes that are supplied by

domestic wells. Three studies, one conducted in Pennsylvania (Swistock, Clemens, Sharpe, & Rummel,

2013), Wisconsin (Knobeloch, Christenson, Anderson, & Gorski, 2013), and Virginia (Pieper, Krometis,

Gallagher, Benham, & Edwards, 2015) serve as the basis of our knowledge of domestic well WLLs data.

Because these three studies provide a backdrop to the research we conducted, they are described in

detail.

1. Sampling Strategy and Sample Analysis

Each of these three studies resulted from collaborations with free and reduced-fee well water

testing programs affiliated with the extension offices of universities and state agencies (**Error! Reference

source not found.**). Two of these studies (Knobeloch et al., 2013; Pieper et al., 2015) used extension

programs where individuals simply submitted samples for lab analyses, and the study authors used

minimal exclusion criteria beyond complete sample information and confirmation that they came from a

private well. By contrast, the third study (Swistock et al., 2013) used a more involved extension program

(the Penn State Master Well Owners Network) that used more senior, trained well owners to help with

selection of wells and to train other wellowners on how to sample and interpret their results.

All three of these studies had homeowners taking their own samples in their home, with instructions provided to them.

TABLE IV- SAMPLING STRATEGY AND INCLUSION CRITERIA OF PRIVATE DOMESTIC WELL STUDIES

Name of Study	Location	Study Design/Sampling Strategy	Population Focuses/Inclusion Criteria
Knobeloch, Christenson, Anderson, & Gorski, 2013	Wisconsin	Study Design: Historic data, 2007-2010 in a voluntary water testing program. Sample Strategy: No imposed design. Considered geographically representative.	Program Focus: Low-income families with pregnant women, or young children. Inclusion Criteria: No inclusion criteria stated by researchers.
Swistock, Clemens, Sharpe, & Rummel, 2013	Pennsylvania	Study Design: Analysis of historic data, 2007. Voluntary water testing program employing a "train the trainers" model. Sample Strategy: Considered geographically representative.	Program Focus: Wells selected to maximize distribution across state. Minimum distance of 1.6 km used between water wells. Inclusion Criteria: No inclusion criteria stated by researchers.
Pieper et al., 2015	Virginia	Study Design: Concurrent with regular data collection (2012-2013). Voluntary water testing program employing a "train the trainers" model. Sample Strategy: No imposed design by researchers. Considered geographically representative.	Program Focus: Any homeowners who wanted to participate. Inclusion Criteria: Data analysis limited to water from private wells

*Lead was evaluated as part of a broader suite of constituents in Swistock et al (2013) and in Knobeloch et al. (2013), as part of a general evaluation of well water quality. Only dissolved lead levels were analyzed for these two studies (*TABLE V- SAMPLE COLLECTION METHODOLOGY AND PARAMETERS OF INTEREST OF PRIVATE DOMESTIC WELL STUDIES

Name of Study	Specific Sampling Goals?	Flushing/Stagnancy Procedures	Controls for Treatment Systems	Form of Lead	Corrosivity-Related Parameters?

| Knobeloch, Christenson, Anderson, & Gorski, 2013 | No primary contaminant of focus in sampling | No stagnant/first draw samples

Water ran for 6 minutes before sampling. | Would take from unsoftened kitchen faucet/pressure tank before system implemented. | Soluble | Other metals as signs of degradation (i.e. Al, Cd, Cr, Cu, Fe, Mn, Ni, Zn), but not used in this fashion |
|---|---|---|---|---|---|
| Swistock, Clemens, Sharpe, & Rummel, 2013 | No primary contaminant of focus in sampling | Collected both first and flushed samples. Unstated stagnancy time. | First draw from kitchen faucet, flushed sample from untreated tap. Network provided training to disengage systems. | Soluble | pH |
| Pieper et al., 2015 | No primary contaminant of focus in sample collection (focus in data analysis though). | First draw sample: 6 hour stagnation.

Collected 250mL "pencil thin flow".

Flushed sample: Ran sample for 5 minutes.

Collected 2 250mL samples and 1 100mL sample | Took sample from non-swivel faucet | Total, Soluble, and Particulate | Total Dissolved Solids, Hardness (Ca, Mg), Cl, SO_4

Other metals as signs of degradation (Ag, Al, Cd, Cr, Cu, Fe, Mn, Zn). |

). Pieper et al., (2015), by contrast, evaluated dissolved, particulate, and total lead to better evaluate possible contributions of lead, as well as any relationships between lead and other metals. In all three studies, water samples were collected by household occupants, who had received sample collection bottles with instructions. With the exception of Pieper et al. (2015), individuals received instructions to disengage or take samples from upstream of any household water treatment system. Additionally, two of them (Pieper et al., 2015; Swistock et al., 2013) asked participants to collect a first draw sample and a running water/flushed tap sample. Nonetheless, only the first draw samples were evaluated for lead in Swistock et al. (2013). Knobeloch et al. (2013) did involve the collection of first draw (stagnant samples); participants only collected flushed samples. Both Knobeloch et al. (2013) and Pieper et al. (2015) looked at metal parameters that could have some utility in terms of evaluating corrosivity, but only Pieper et al specifically looked at these from that perspective.

2. <u>Findings of the Three Studies</u>

The results of these three studies are summarized in TABLE *VI*. Two of the studies with first

draw samples had exceedances of the EPA action level of 15 µg/L, but Pieper, et al. (2015) found almost

twice as many exceedances with a much larger sample size counting particulate as well as dissolved

lead. Only Pieper et al. (2015) indicated the overall distribution of lead values, indicating that 73% were

below 10µg/L and lower. For the second draw, again only two of these studies properly analyzed

flushed samples, but they are consistent with each other in terms of the percent of wells still exceeding

the action level. The actual measurements, however, appear to be quite different. Pieper, et al (2016)

reported results on a log scale, indicating that about 90% of results were under 2.6 µg/L on a logarithmic

scale, with an absolute maximum magnitude of 405 µg/L. This is in stark contrast to Knobeloch et al.

(2013), which reported a much higher median of 9 µg/L and a maximum reported value of 2100 µg/L. It

is also worth pointing out that, for Swistock et al. (2013), samples were only sampled for lead in the

2007 analyses.

TABLE V- SAMPLE COLLECTION METHODOLOGY AND PARAMETERS OF INTEREST OF PRIVATE DOMESTIC
WELL STUDIES

Name of Study	Specific Sampling Goals?	Flushing/Stagnancy Procedures	Controls for Treatment Systems	Form of Lead	Corrosivity-Related Parameters?
Knobeloch, Christenson, Anderson, & Gorski, 2013	No primary contaminant of focus in sampling	No stagnant/first draw samples Water ran for 6 minutes before sampling.	Would take from unsoftened kitchen faucet/pressure tank before system implemented.	Soluble	Other metals as signs of degradation (i.e. Al, Cd, Cr, Cu, Fe, Mn, Ni, Zn), but not used in this fashion
Swistock, Clemens, Sharpe, & Rummel, 2013	No primary contaminant of focus in sampling	Collected both first and flushed samples. Unstated stagnancy time.	First draw from kitchen faucet, flushed sample from untreated tap. Network provided training to disengage systems.	Soluble	pH

| Pieper et al., 2015 | No primary contaminant of focus in sample collection (focus in data analysis though). | First draw sample: 6 hour stagnation.

Collected 250mL "pencil thin flow".

Flushed sample: Ran sample for 5 minutes.

Collected 2 250mL samples and 1 100mL sample | Took sample from non-swivel faucet | Total, Soluble, and Particulate | Total Dissolved Solids, Hardness (Ca, Mg), Cl, SO_4

Other metals as signs of degradation (Ag, Al, Cd, Cr, Cu, Fe, Mn, Zn). |

TABLE VI- WLLS OF PRVIATE DOMESTIC WELL STUDIES

Study	# Private Well Samples Analyzed for Lead	% First Draw Exceeding EPA Action Level	First Draw Distribution	Second Draw/Flushed-Exceeding Action Level	Second Draw/Flushed Distribution
Knobeloch, Christenson, Anderson, & Gorski, 2013	3,868	Not Applicable	Not Applicable	1.80%	Median (µg/L): 9 Max: 2100
Swistock, Clemens, Sharpe, & Rummel, 2013	251[1]	12%	Not Available	Not Applicable	Not Applicable
Pieper et al., 2015	2,029	19%	Mean: 22 µg/L Median: 4 µg/L	0.70%	90% <2.6 µg/L

[1]Total in study was 701, but only 251 were evaluated for lead.

3. Limitations of the Studies

While these three studies are the most relevant evaluations of lead in private well water, they suffer from a variety of shortcomings (TABLE *VII*). Corrosivity's effect on lead level was not fully measured in any of the studies. In one study, corrosivity was indirectly addressed based on the finding of statistically significant associations between lead, high pH, and self-reported plumbing materials. (Pieper et al., 2015) utilized pH and correlations between lead with zinc, copper, and nickel to evaluate internal corrosion of particular parts (e.g. brass alloys). Additionally, this study addressed corrosivity by looking at the odds of participants observing or noticing specific visual and aesthetic features of their

water and plumbing fixtures and having an elevated water lead level above the action level. However, corrosivity was not explicitly quantified using a standardized metric.

The findings of the three studies are likely specific to their respective geologic setting and potentially groundwater influencing-land use regimes and may have limited relevance to the Illinois environment. Knobeloch, et al. (2013), whose Wisconsin setting most closely resembles Illinois's, did not mention the influence of geology. Swistock et al., (2013) did take geology into account, using the Pennsylvania geologic regions as a categorical predictor in logistic regression and analysis of covariance (ANCOVA) models, but did not report the results of these analyses. Pieper et al. (2015) itself did not discuss geology, but a follow-up study using the same data did seriously evaluate the influence of geologic setting, lead levels, and plumbing using chi-square analyses (Pieper, Krometis, Benham, & Gallagher, 2016). The follow-up study did try to address some of this variation in terms of geology. However, the setting for this latter study (Virginia) is more similar to Swistock, et al. (2013), and does not properly describe the geologic setting of areas like Illinois. The largest proportion of Virginia's geological regions has fractured crystalline bedrock aquifers composed of less reactive igneous and metamorphic rock. As a result, groundwater in these locations is more acidic, has lower conductance and less buffering capacity. The second largest geologic region, Valley and Ridge, is composed of carbonate aquifers that may influence groundwater alkalinity (Pieper, Krometis, Benham, et al., 2016). By contrast, Illinois is majority composed of carbonate and sand and gravel aquifers and have a greater influence on the alkalinity (Illinois State Water Survey, n.d.). Accordingly, the groundwater in Illinois generally has higher pH levels closer to neutral (W. Kelly, personal communication).   A shortcoming found in all three of these studies is the lack of in person training, which is attributable to the researchers using data collected from pre-existing state and university extension programs rather than defining their training program. Since training could not be performed in the place of sampling, there may be unexplained error that could not be controlled in the results presented here.   Finally, only one

study (Pieper et al., 2015) analyzed particulate, soluble, and total lead; the other studies employ the

traditional soluble lead metric and subsequently may be underestimating the full lead exposure.

TABLE VII- STUDY LIMITATIONS OF THE PRIVATE DOMESTIC WELL STUDIES

Study	Corrosivity	Participant Training	Geology	Forms of Lead Evaluated
Knobeloch, Christenson, Anderson, & Gorski, 2013	None	Only instructions provided	Not addressed	Soluble
Swistock, Clemens, Sharpe, & Rummel, 2013	Just pH. Only addressed situations with acidic groundwater	Only instructions provided	Uses Pennsylvania geologic regions, used in logistic regression and ANCOVA models. Outputs are not reported beyond parameter significance.	Soluble
Pieper et al., 2015	pH and correlations between lead and copper, zinc, and iron as indicator of corroded piping/fixtures/solder. Does not address situations of high pH	Only instructions provided	Not addressed in this study, but addressed subsequently in Pieper et al 2016, very specifically tailored to Virginia geology.	Total, Soluble and Particulate

E. Corrosivity and Groundwater

1. Metrics of Corrosivity

One metric of corrosivity is the Larson-Skold index. This index was developed particularly using

Great Lakes water using in-situ measurements of corrosion of steel lines (Larson & Skold, 1958). It uses

the concentrations of chloride and sulfate which can interrupt the formation of natural carbonate films

and scales.   The equation for deriving it is the following:

$$L \,\&\, SkI = \frac{(epm\ Cl^- + epm\ SO_4{}^{2-})}{(epm\ HCO_3{}^- + epm\ CO_3{}^{2-})}$$

Larson Skold Index (LSI) evaluates at the equivalents per million (EPM) of each chloride (Cl^-) and sulfate (SO_4^{-2}) divided by the alkalinity (measured as forms of available carbonate). When resulting values of the LSI are <0.8, it is assumed that chloride and sulfate are not present in large enough concentrations to hinder scale development. Values of 0.8 to 1.2 indicate that scales may not develop and that water may be corrosive. Values greater than 1.2 indicate that elevated rates of corrosion are expected to occur.

Another metric of corrosivity is the chloride to sulfate mass ratio (CSMR). This metric more qualitatively measures corrosion rate particularly related to galvanic corrosion. Results of studies from Nguyen, Stone, and Edwards, 2011 indicated that a CSMR <0.2 is of no concern, but higher (0.2-0.5) is significant, and even higher (>0.5) with low alkalinity (<50 mg/L $CaCO_3$) is of serious concern.

2. Geologic and Anthropogenic Influences on Corrosivity and Corrosivity-Related Species

Groundwater chemistry, and subsequently the corrosivity of groundwater, is a product of its geologic setting and land use. There are two major types of aquifers in Illinois. The first are shallow surface aquifers, composed of unconsolidated deposits of sand and gravel in the form of glacial till (deposited material from historic glacial activity or alongside stream valleys). The largest unconsolidated aquifers are found in northeastern and central Illinois. The other type of aquifer, porous and permeable bedrock, is primarily found in the northern third of the state, though there are isolated pockets the in southern and western portions of Illinois as well. Many bedrock aquifers in the southern two-thirds of the state are quite saline and undrinkable. Bedrock aquifers in Illinois are typically sandstones and carbonates (limestone and dolomite) (Samuel V. Panno & Hackley, 2010, p. 19).

Groundwater geochemical parameters are determined in part by the locations in which water percolates into the ground. Precipitation can receive ions from seawater aerosols, airborne dust, and

from different anthropogenic inputs, such as traffic exhaust, and industrial emissions. However,

groundwater chemistry is mostly determined by soluble minerals and ions picked up as it flows through

the subsurface. Groundwater can become saturated with calcium, magnesium, bicarbonate, and other

ions as it percolates downward, and its TDS tends to increase with distance. Therefore, shallower

surface aquifers tend to have fresher water with lower TDS. Elevated TDS and chloride levels in

groundwater, such as often found in deeper bedrock layers like the underlying sedimentary Illinois

Basin, are naturally occurring (Samuel V. Panno & Hackley, 2010). However, even with historical saline

influences, most aquifers in Illinois have low chloride concentrations. When looking at the chloride

concentrations of shallow northern Illinois groundwater, Panno et al (2006) found that natural

background chloride levels ranged from under 1 mg/L to 15 mg/L. However, chloride levels can be

influenced by anthropogenic inputs, such as from rock salt applied to melt snow and ice on roads (Kelly,

2008). Thus, shallow aquifers with chloride concentrations in excess of 15 mg/L are likely to be

impacted by anthropogenic sources. Sulfate is naturally occurring in Illinois groundwater, usually no

more than tens of miligrams per liter. However, concentrations upwards of 2,000 mg/L in the Cambrian

and Ordovician aquifers in northeastern Illinois, as well as in portions of the Mahomet aquifer in east-

central Illinois (Samuel V. Panno & Hackley, 2010). Construction can disturb and unearth sulfide rich

minerals to air exposure, resulting in oxidation reactions that yield sulfate and sulfuric acid (Wagner,

Fanning, Foss, Patterson, & Snow, 1982). Active and inactive or reclaimed coal mines, particularly in the

southern part of the state, similarly expose sulfur-containing minerals, reacting with precipitation to

produce sulfuric acid. This can subsequently percolate into groundwater, raising the sulfate and

lowering the pH (Kemmis, Bauer, & Lasemi, 2010).

F. **Corrosive Water and Domestic Well Water Systems**

1. **Sources of Lead in Wells and Premise Piping**

There are various components of well and domestic plumbing systems that corrosive

groundwater can interact with and subsequently become contaminated with dissolved and particulate

lead (

Figure 1). These include parts of the pump, the wellhead itself, the packer, the well screen, the well

casing, submersible water pumps, pipes, pitless adapters, the pressure tank, faucets, and various fittings

and joints and alloys (Pell & Schneyer, 2016). If the well or home are old enough, the plumbing may be

composed entirely of lead. Use of lead in new service lines and plumbing was banned in 1986, and for

that reason, homes, wells, and plumbing systems built since then should contain significantly less lead

than older materials.

Solder alloys containing up to 40-50% lead were also extensively used until being banned in

1986 to join plumbing materials with a water tight seal (Triantafyllidou & Edwards, 2012). In particular,

lead solder has been demonstrated in lab analyses and case studies to corrode and contribute to

particulate lead exposures (Triantafyllidou, Parks, & Edwards, 2017). Many components like valves,

faucets, and fittings inside the home, as well as submersible pumps before 1995, may contain brass,

another potential source of lead. Both lead solder and brasses connected to copper pipes can undergo

galvanic corrosion at high rates, particularly in the presence of a high CSMR (Nguyen, Stone, & Edwards,

2011).

The impacts of brass fixtures may especially contribute to lead exposure in a given home.

Kimbrough (2001) looked at a natural experiment in municipal water systems, where a new tract of

homes was built along with a new public water system to service it. The higher order street mains for

this new tract were made of ductile iron, the service lines were made of copper, and the meters had

"lead-free" brass components. Additionally, the plumbing systems connecting the meter to the tap were made entirely of plastic with some organolead components. It was hypothesized that the major sources of lead would come from the brasses in the interior fixtures. Samples from the new plastic plumbing homes were compared to previously sampled, traditionally plumbed homes. This control population had no lead service lines or plumbing, just lead-soldered copper pipes. Concentrations of copper, lead, nickel, and zinc (brass corrosion-indicator elements) were higher in the newer plastic homes. Additionally, samples that were high in zinc were also likely to be higher in lead and nickel, which corresponds with the mechanism of dezincification common to brass corrosion.

Galvanized steel pipe, which has a protective zinc layer, can also contain lead impurities (Triantafyllidou & Edwards, 2012). Finally, the chemical scales and rusts that form on iron and other types of pipes downstream from a lead source may adsorb lead, releasing it much later after physical or chemical disturbances. This means that lead may continue being released even after the upstream lead-containing elements of the plumbing system have been removed.

In a recent analysis, Pieper, Nystrom et al. (2018) extensively sampled 15 homes in Macon County in North Carolina and analyzed lead and metals concentrations in each of the first draw and flushed samples. Samples were taken directly at the wellhead to avoid the influence of premise plumbing, and all of them contained detectable lead, with ten of them exceeding the action level by a factor of 1.2-116. All the wells were confirmed to have had brass fittings at the sampling outlet, and 13 of them had galvanized iron components at the well head. The presence of brass and iron in the fittings was consistent with the observation of significant correlations between particulate lead levels and zinc ($r=0.55$), copper ($r=0.71$), and iron ($r=0.50$), respectively.

It is important to note that the ongoing presence of leaded well components may be the result of regulatory ambiguity and oversight at the state level. Pieper, Krometis, and Edwards (2016) discuss

this in evaluations of the National Science Foundation International/American National Standards Institute's lead-free standards. They note that the 1986 Lead Ban used the language of "any plumbing… which is connected to a public water system", which may have excluded private wells. Well components appear to have had the ban enforced in 1995 due to concern of submersible pump leaching. While this has been resolved, there are still ambiguities in the regulations such that state agency oversight may contribute to the continued use of leaded component use in private well systems (Pieper, Krometis, & Edwards, 2016).

2.   <u>Lead Levels as a Product of Geologic Factors and Premise Piping/Well Construction</u>

While there is literature on the impact of geologic setting on groundwater quality and drinking water, and some literature on the impact of well construction and premise plumbing on drinking water quality, there is almost no literature about the combined impact on drinking water driven by both geologic setting and plumbing and well design. A single study conducted in Virginia addressed this, using data from the aforementioned Virginia extension program (Pieper, Krometis, Benham, et al., 2016). As previously mentioned in greater detail, stagnant water samples were collected from point of use. To classify geology, the investigators looked to see if a county was labelled 75% or greater of single state geologic region, and used wells that fell within those counties. There were 3 major geologic units: Coastal Plains, Blue Ridge/Piedmont, and Valley and Ridge. The Coastal Plains region consists of unconsolidated and semiconsolidated aquifers, and the groundwater in this region flows between sediment grains. There is heterogeneity in terms of groundwater quality and may be impacted by saltwater intrusion.

Figure 1- Infographic of lead-bearing components in a private domestic well system (Pell & Schneyer, 2016).

Ground to tap: sources of lead

Lead in tap water from houses that use private wells almost always derives from components of the well or residential plumbing, according to Bryan Swistock, a researcher with Penn State Extension. As in Flint, corrosive water from wells leaches lead from plumbing parts it comes in contact with, from the well screen to the faucet.

Plumbing
Pipes and fixtures can contain lead and joints can include lead solder; until January 2014, plumbing fixtures designated as lead-free can contain up to 8 percent lead.

Well cap

GROUND SURFACE

Pitless adapter
Components may contain brass that includes lead

Pressure tank

WATER TABLE

Pipes
Even new pipes made of galvanized steel can contain lead that could be mobilized by aggressive water.

Corrosive water
Ground water that is acidic or contains high levels of salt can erode plumbing and well components, releasing lead into the tap water.

Submersible water pump
The pump can use brass valves that contain lead.

Well casing
Metal in the casing protecting the pipe can contain lead.

Well screen

Source: Reuters   Diagram not to scale.

Blue Ridge and Piedmont are two different regions lumped together with different topography, but similarly have fractured crystalline bedrock aquifers with overburden thicknesses. Groundwater present here had a low pH and has low conductivity (which presumably is in line with low TDS as well). Valley and Ridge has a mountainous topology that was more conducive to deeper drilled wells. The groundwater in this region dissolves the carbonate-rich aquifers, which subsequently raises the groundwater's pH. It also had a greater amount of interconnected solution channels, which can increase groundwater penetration. In this study, well design was analyzed in terms of whether wells were drilled or bored. Major differences between drilled and bored wells are the differences in depth (drilled wells

are much deeper) and that there is a discontinuous casing around bored wells as opposed to the fully cased drilled wells. The researchers then conducted chi-squared tests using three-dimensional contingency tables.

The authors found that WLLs from wells did differ with geology, with the Blue Ridge-Piedmont area having higher lead concentrations than Valley and Ridge, which were significantly higher than the Coastal Plain region. Dug/bored wells had higher lead exceedances than their drilled counterparts. The Coastal Plain Region had fewer drilled wells exceeding the action level than expected, but had greater than expected bored wells with higher lead concentrations. This was attributed to lower observed pH levels, presumably because the groundwater spent less time percolating and dissolving minerals that could increase its buffering capacity. The Blue Ridge-Piedmont region had higher than expected numbers of both drilled and bored wells exceeding the standard, attributed to a lack of geochemical buffering capacity and fractured groundwater flow. Finally, the Valley-Ridge region had a more complex geological setting, with lower than expected counts of drilled wells, but median lead concentrations significantly higher than Coastal Plain, but lower than Blue Ridge-Piedmont. This variation was attributed to the known buffering capacity of the Valley Ridge region (median pH= 7.3) when compared to the Blue Ridge-Piedmont region, but the authors also noted that the presence of solution channels enables acidic groundwater to more easily penetrate the ground and flow into wells than in Coastal Plain, which had very acidic water.

The only other study that mentions both geology and well design is the previously mentioned study conducted in Pennsylvania (Swistock et al., 2013). As part of this study, the researchers looked at the association of various categorical variables related to well, geology, location, and plumbing to lead levels using ANCOVA and logistic regression. The researchers classified the bedrock geology as carbonate, inter-bedded sedimentary, sandstone/shale, or conglomerate. Well characteristics were summarized by a "well score" of good well construction practices (i.e. grouting), as well as depth and

well casing material. Plumbing type (plastic vs metal), as well as plumbing age (pre/post-1991) were

included as well in these models. The authors of this study did not present the actual results of these

model runs, but indicated that statistically significant models explaining lead concentration variation

could be linked to geology and plumbing type. While the data were not shared explicitly, the authors

noted that 70% of homes with elevated water lead levels also had copper plumbing systems, and 93% of

these homes also had acidic water. The authors, however, did not provide proper contextual data about

the percent of copper plumbing systems overall or in non-elevated WLL homes, which limits the utility

of these data.

Neither of these studies, however, evaluate enough information to relate mechanistically or

statistically how both geologic setting and well/plumbing systems contribute to lead levels. Pieper et al.

(2015) did not evaluate the impacts of premise plumbing in relation to wells or try to gather more

information about wells and well components. Swistock et al. (2013) by contrast, did include

information about premise plumbing and age, as well as well design and age. Both of these studies,

however, mostly rely on categorical metrics to broadly classify geology rather than taking into account

local factors. Most importantly, both of these studies did not fully evaluate corrosivity. In both cases,

the assumption is made that corrosivity is solely a function of low pH, and that pH is the main parameter

that relates to corrosivity. While low pH water is often corrosive, this does not take into account the

many other factors that influence the corrosivity content of groundwater, as well as the context-

dependent interactions of pH with a given setting.

     G. Potential for Intervention

          1. Well Remediation and Filtration Technologies

Lead exposure from private wells is a solvable problem. While its extent in Illinois specifically is

unknown, the direct exposure can be attenuated and minimized through technological and behavioral

interventions. The most direct method of intervention is to remediate older wells and plumbing with

lead problems, or to replace all lead-bearing components with lead-free ones. As has been mentioned,

the most likely sources of lead in wells are the galvanized iron well casings, galvanized iron and brass

well parts, and scales and sediments with adsorbed lead (Pieper, Nystrom, et al., 2018). This approach is

the most comprehensive one, and would require the use of well cameras to ensure that scales and

sediment were removed. At the same time, this approach is quite difficult and expensive, especially to

locate which components are being corroded and releasing lead.   It would not be a standardized

process and would be driven by the home plumbing system, geologic setting, state of the groundwater,

and the well structure, and, while there would be some home assistance programs, much of the cost

would be the responsibility of the homeowner because of the lack of present regulatory mandate or

more robust government support programs. While this approach is the most extensive one, it is the

most burdensome one for homeowners, and may not necessarily be realistic.

Filtration devices and apparatuses, therefore, are a major method of reducing waterborne lead

exposures (Minnesota Department of Health, 2014, n.d.). Both POE and POU devices may be employed

for this purpose. Since lead in private wells is a product of corrosive groundwater, POE devices can

control for some of the properties that contribute to corrosivity as groundwater enters the household

plumbing system from the well, before the pressure tank. Acid water neutralizing filters, for example,

can raise the pH by directing water flow through carbonate-rich media (i.e. calcium carbonate, marble

chips). However, these require periodic maintenance. Additionally, they often need to be used in

tandem with a water softener since they raise the alkalinity, and may trap particles and oxidized metals

and further require maintenance unless a sediment filter is also installed, increasing overall costs.

Another POE device that can control corrosive conditions use sodium hydroxide or soda ash chemical

pumps. These are also quite effective at raising the pH, but are similarly very maintenance intensive, as

the chemical pumps need to constantly be refilled. POE devices are most useful when the most

problematic lead-bearing components are found in the household plumbing. If corrosive water has

already come into contact with components in the well, then lead will still come out at the point of use.

By contrast, POU devices are at the tap proper and are used to filter out particular contaminants

at the tap. One such system is an activated carbon filter, whose carbon particles contain many tiny

pores, allowing water through and adsorbing organic and choice non-organic pollutants. These have to

be specifically designed with lead in mind and can only process a small amount of water, requiring

frequent replacement. Another prominent POU method is reverse osmosis, where water is forced

through a small semipermeable membrane, leaving behind all impurities. This procedure is incredibly

energy and water intensive, as only 10 to 30% of incoming water is produced as drinkable water

(Masarik, 2007). Additionally, it requires constant maintenance, as the membrane wears down

overtime. The third major POU apparatus is the use of distillation units, which can boil away incoming

water and then cool the resulting steam to reconstitute it, leaving the contaminants behind. This

process, like reverse osmosis, is also quite energy intensive. It also may need to be coupled with a water

softener, since minerals and other impurities tend to accumulate in the boiling chamber and hard water

particularly can clog it. Distilled water is also corrosive and needs to be handled carefully and placed in

particular containers, to ensure it does not dissolve pollutants. While not addressing the actual cause of

high water lead levels, POU devices can be used to reduce exposures from well components if the well is

inaccessible or unable to be remediated, and can reduce exposure from lead-containing components in

premise piping. However, POU devices are downstream and do not address the source conditions or

components, but rather mitigate exposure to them.

2. Flushing

Flushing has been explored as a low cost option for mitigating water lead exposure. The official

CDC recommendation for private wells is to flush each tap before use on cold for 1-2 minutes for any

sink that has been stagnant for 6 hours or more (CDC, 2015). From our model studies, this approach can

be fairly effective. In Knobeloch et al. (2013), only 1.8% of flushed samples from wells (n=66) exceeded

the EPA action level as part of a flushed sample. Similarly, in the Virginia study (Pieper et al., 2015),

0.70% of flushed samples from wells (n=15) were above the threshold. However, 2% of all samples

(n=46%) increased in the flushed sample in terms of concentration. These increases were modest

(mean= 5.2 µg/L, median= 1.0 µg/L), with 8 homes having concentration increases much greater than 5.

The efficacy of flushing appears to be dependent on the components of the well, plumbing

system, and fixtures, and how they are corroding. This was demonstrated in the North Carolina case

study (Pieper, Nystrom, et al., 2018), where first and second draw samples were taken on a small

amount of wells (n=20), followed by 1 L samples at different flushing intervals (1, 2, 3, 5, 10, 15

minutes). There were clear declines between first and second draws, and the median water lead levels

across all flushing intervals were lower than the first draw sample. During the flushing exercise, there

were 6 wells with sporadic lead spikes (1.0-7.2 µg/L), 5 of which had produced non-detectable samples

in previous flushes. Measured lead in these samples was all particulate and significantly correlated with

zinc and iron levels, indicating that the lead source was most likely galvanized components of the

wellhead and the well itself. One well had detectable lead in particulate form for all flushes and had all

of its lead components removed previously, indicating all detectable lead were deposits produced by old

lead well and plumbing components. The inconsistency and site specific nature of flushing approaches

and flushing guidelines is corroborated in a study with municipal systems in New Orleans, using a much

larger sample size (Katner et al., 2018). Participants took water samples over the course of multiple

flushing intervals (first draw, 30-45 s, 2.5-3 min, and 5.5-6 min) found that even when lead

concentrations declined at the 6 minute flush, 52% still had detectable levels of lead (defined here as ≥ 1

ppb). Particulate lead may be picked up from premise plumbing in response to excessive flow, but its

mechanism of release is inconsistent and requires further study. Flushing as a practice can overall

reduce water lead exposure, but it is less reliable when compared to the use of filtration systems or well remediation.

3. Wellowner Knowledge and Perception of Risk

The liability and responsibility of private well water quality and safety rests solely on each individual wellowner, meaning that any protective or reparative measures undertaken are limited by their own knowledge of well maintenance. Based on a report using Wisconsin BRFSS data (Knobeloch, 2010), it appears that there is an underperception of the risks associated with private drinking water wells, particularly of issues involving water lead. This problem is ripe for an intervention, as even simply raising the awareness of well owners is effective in getting them to pursue mitigation options. In Knobeloch et al. (2013), participants whose wells had coliforms exceedances were mailed a follow up survey 6 months after the study (n=60). They indicated that none of them tried to avoid lead before the study, but after testing, 75% began taking measures to reduce exposures. Similarly, well owner perception of signs of well/plumbing corrosion could predict whether there would be a greater water lead concentration. In Pieper, et al (2015), homeowners that identified different signs relating to corrosion (blue-green staining on fixtures, metallic flavor to water, obvious signs of corrosion) had 1.7 to 2.8 times greater odds of having elevated water lead concentrations than homeowners who did not see these indicators. While perception as a metric is related to various factors like education and income, this suggests that empowering homeowners by teaching them how to maintain and see the signs of well corrosivity can push them to reduce their own exposures.

III.   INFORMATION/PRACTICE GAP AND RESEARCH OBJECTIVES

There is little complete information on water quality in private domestic wells, particularly regarding water lead levels in the state of Illinois. While there have been previous lead well studies (Knobeloch et al., 2013; Pieper et al., 2015; Swistock et al., 2013), these by and large have not evaluated corrosivity appropriately, which would dictate forms of lead present in the water and appropriate prevention and remediation efforts. These studies are also plagued with methodological inconsistency in terms of characterizing both premise plumbing and domestic well water via stagnant, first draw and flushed, last draw lead samples. Two of three of these major studies did not sufficiently look at all bioavailable forms of lead, and may not necessarily be applicable to the specific hydrogeological regime of Illinois. Better understanding this phenomenon has direct application at the public health level, since health departments in rural areas do not have cost effective, scientifically derived decision rules for understanding wells and homes that are higher at risk for lead contamination based on corrosivity information.

Thus, our study aimed to better understand the influence of corrosivity and well and premise plumbing quality on lead levels of private domestic well-supplied homes, particularly with regard to developing screening decision rules for local health departments (LHDs). We aim to properly describe corrosivity mechanisms, capturing both impacts from private domestic wells and premise plumbing, and quantify total lead and partition between dissolved and particulate lead in Illinois private domestic wells.

Our research objectives were as follows:

- To evaluate corrosivity and housing age as predictors of total water lead levels in private well-supplied homes in rural Illinois.

- We expected that more corrosive water, interacting with older homes and wells, would produce higher lead levels at the tap

- There would be a differentiation of the forms of lead based on corrosivity and housing characteristics

IV.   METHODS

A. Study Design

We conducted a cross-sectional pilot study to evaluate total available lead and other relevant metals, corrosivity metrics, well and housing information, and demographics. Phase I consisted of initial recruitment into the study and sampling of homes, while Phase II would consist of a closer evaluation of homes with elevated levels by the Illinois State Water Survey.

B. Study Overview
The following schematic summarizes major aspects of the study:

Figure 2- Summary of major study elements
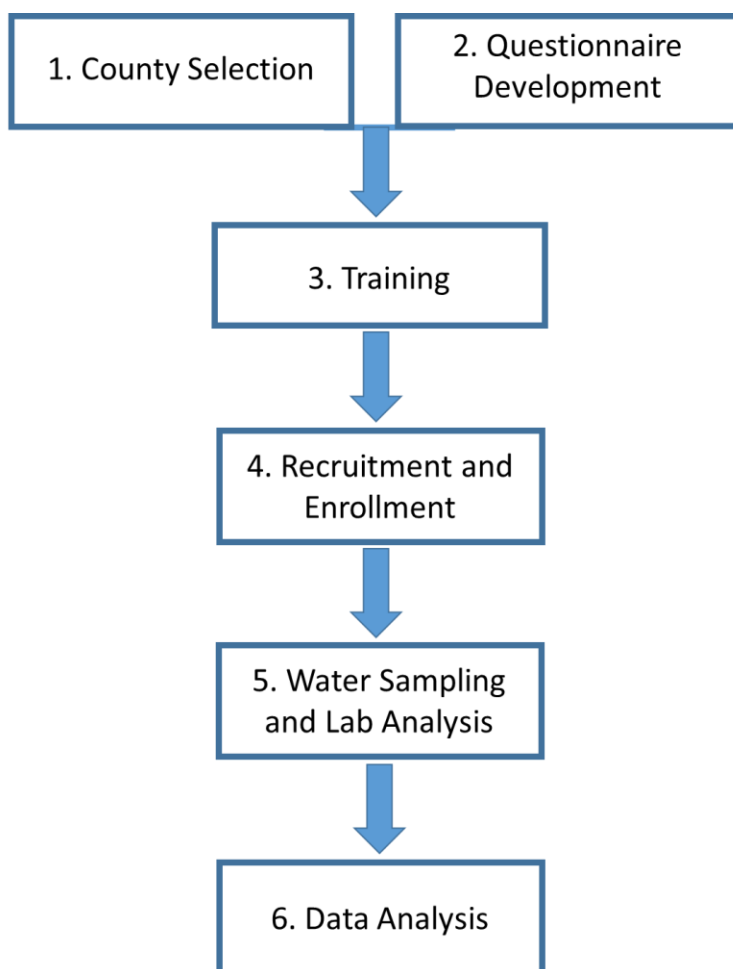
1. **County Selection and Training (Steps 1, 3)**

We looked for 3 prospective counties and county health departments that were in the northern, center and southern parts of the state, to reflect potential geographic variability. To start scoping out counties, we began by looking at publicly available data on blood lead levels, housing stock, and corrosivity. Blood lead level data were compiled from the Illinois Lead Program 2014 Annual Surveillance Report. Corrosivity of wells per county were evaluated by calculating high Larson-Skold Indices using data from the Illinois State Water Survey Domestic Well Database. We also looked at percentages of pre-1950s housing using data from the United States 2000 Census. All of these indicators were used to isolate which counties were preferentially contacted. Ultimately, though, we selected county health departments that were the most interested in participating in the research rather than necessarily counties that best fit our demographic, hydrogeological, and housing criteria.

We selected Kane County (North), Peoria County (Central), and Jackson County (South). We discussed and shared with the points of contact at each health department our study materials and procedures to ensure that they were compatible with the department's policies, and later went out to conduct in-person research protocol trainings at each location for all involved staff. Through this process, we were able to get constructive feedback in terms of study flow and materials. As an example, we had our questionnaire translated into Spanish in order to accommodate the large Spanish speaking population in Kane County.

2. **Questionnaire Development (Step 2)**

Initially, we attempted to find already validated survey instruments that used semi-quantitative methods of evaluating water consumption. We looked through national surveys (i.e. American Community Survey (ACS) NHANES, BRFSS), looking particularly for sections that collected information on water consumption and use, perceptions of water quality, and information on housing and plumbing quality and characteristics. We also looked for other studies that used survey instruments evaluating

different contexts of water consumption, exposure, and quality perception. We were unable to find a fully validated questionnaire for our purposes, since most questionnaires that do quantitatively evaluate water consumption are Food Frequency Questionnaires (FFQ), which require computer or in-person administration and are more appropriate for dietary and nutrition-based studies. Some questionnaires asked if an individual did or did not consume well water, but gave no indication about the amount consumed. Ultimately, we created an composite questionnaire using questions from multiple sources (Bureau, 2016; Erinosho et al., 2015; Hargrove, Juárez-Carillo, & Korc, 2015; Jones et al., 2006; McLeod, Bharadwaj, & Waldner, 2014; Merkel, Bicking, & Sekhar, 2012; Onufrak, Park, Sharkey, & Sherry, 2014), and further refined it from there. This questionnaire was not validated.

3. Recruitment and Enrollment of Participants (Step 4)

We received IRB approval to start recruitment in Kane and Jackson County in June and July 2018, and approval for Peoria County in October 2018. Each local health department (LHD) disseminated flyers, put out press releases and used social media to promote the study. They also conducted public outreach and attended meetings as needed. When prospective participants would call in, IRB-approved LHD staff would check verify their county of residence, whether they got their water from a domestic well, the estimated age of their home, and their willingness to have project staff conduct a home visit.

After checking eligibility of individual households, members from each LHD contacted eligible participants by phone or email (based on the preferred communication noted by participants on the eligibility screen) to schedule water sample collection training and to provide sampling materials and the study questionnaire. This could be arranged at the participant's home, or at the LHD. The LHD staff member obtained written informed consent from the participant before demonstrating how to properly take water samples. The LHD staff member then left paper questionnaires and water sample collection

and shipping supplies (i.e. bottles, shipping stickers). Water sampling methodology is further described in the Water Sampling and Laboratory Analysis section below.

We intended to oversample older homes. After the first 20 participants in county were enrolled, we restricted eligibility based on the age of housing. Specifically, we checked if the homes were older than 1978, when Consumer Protection Safety Commission implemented regulation banning lead paint and lead paint-containing products (16 CFR § 1303.1- Scope and application., 1977). Our initial goal was to get about 15-20 homes within each quadrant of every county, with 15-16 older homes and 2-3 new homes in each quadrant. We hoped to have 85% of all participating households to be older housing stock.

4. <u>Water Sampling and Laboratory Analysis (Step 5)</u>

We followed USEPA's guidance for collecting samples to comply with the Lead and Copper Rule (United States Environmental Protection Agency, 2008).   We instructed participants through the home visit and through training videos made available at the Private Well Class website (http://privatewellclass.org/lead-sampling), a program developed to educate private well owners. Two labeled 1-L high density polyethylene (HDPE) bottles were distributed to the residents. A "first-draw" sample for Pb was collected in one of the 1-L bottles from a cold water kitchen or frequently used bathroom tap, where the water had lain stagnant in the pipes for at least six hours (i.e., no flushing, showering, etc.). The second 1-L bottle was used for a 7th L "last-draw" Pb sample from the same tap. This "last-draw" sample was also used for anion and alkalinity analysis. After filling the first bottle, the resident filled the second bottle five times, discarding the water each time, before filling the bottle a sixth time and save that sample. The participant then shipped the samples to the Illinois State Water Survey (ISWS) by the resident in a pre-addressed and post-paid insulated shipping container, which included a freezer pack. Samples were shipped so that they reached the ISWS within 3 days of sample collection.

Samples were analyzed at the ISWS for total lead, pH, alkalinity, and anions (chloride and sulfate). For the lead analyses, the method was based on US EPA Method 200.9 (Environmental Monitoring Systems Laboratory, 1996). The minimum detection limit and limit of quantification for this method was 0.76 µg/L for our data. Samples were preserved with 0.2% nitric acid and subsequently digested in 3% nitric acid before being analyzed. Measurements were made using an Agilent Technologies 240Z Graphite Furnace Atomic Absorption Spectrometer, with Zeeman background correction, a PSD 120 Programmable Sample Dispenser, and SpectAA software. Samples with the highest total Pb contents were filtered through a 0.45 µm filter for dissolved Pb analysis. The analytical procedure was the same as for total Pb.

Anions (sulfate, chloride) were analyzed using US EPA Method 300.0 (Environmental Monitoring Systems Laboratory, 1993, p. 0). Only sulfate samples required preservation via cooling to 4°C. The method detection limit for chloride was 0.16 mg/L and 0.21 mg/L for sulfate. Both species were analyzed using a Dionex ICS-5000 ion chromatograph (25-microliter injection loop) with a conductivity detector, an AS-DV automated sampler, and an AERS 500 eluent suppressor. Separations were carried out isocratically on an IonPac AS14 analytical column, with an AG14 guard column, using a mixture of 1.0 mM sodium bicarbonate and 3.5 mM sodium carbonate as an eluent. Chromeleon software was used to collect and process the data.

Alkalinity measured in the form of $CaCO_3$ was determined using Standard Method S2320B (American Public Health Association, American Water Works Association, & Water Environment Federation, 1999). Laboratory pH measurements were analyzed using US EPA Method 150.1 (United States Environmental Protection Agency, 1983). For both of these parameters, the pH of each sample was determined by potentiometric measurement of the hydrogen activity using a combination electrode and a pH meter. The procedure was automated, using a Mettler Toledo T70 titrator, a Mettler DGi111-

SC combined glass pH electrode, and a Rondo autosampler. LabX light software was used to collect and

process the data.

5. Data Analysis (Step 6)

Questionnaire data were input and managed in Microsoft Access form, with appropriate quality

assurance/quality control (QA/QC) procedures to ensure the accuracy of the data. We specifically used

duplicate data entry to ensure data quality, specifically extracting data on age of housing. Water

analytical data were processed to allow for comparisons between first draw and last draw lead samples

by household sampled, and were merged together with corresponding results for the questionnaire. For

all chemistry data with non-detect results, we used single imputation methods, using a standard

environmental data convention as follows:

$$\frac{Sample\ QuantificationLimit}{\sqrt{2}}$$

For each sample, we calculated both the Larson-Skold Indices and the CSMR. The Larson Skold Index

was calculated using the following equation:

$$L\ \&\ SkI = \frac{(epm\ Cl^- + epm\ SO_4{}^{2-})}{(epm\ CaCO_3)}$$

For samples in which dissolved Pb concentrations were determined, particulate lead was calculated by

subtracting dissolved lead concentration from total lead concentration.

All analyses were conducted in SAS 9.4 (SAS Institute, Cary NC). Univariate analyses were

conducted for WLLs, geological parameters, corrosivity (both Larson Skold and CSMR), and age of

housing. We first checked for normality of these using the Shapiro-Wilk test. Housing age was normally

distributed, and both corrosivity metrics became normally distributed after applying log

transformations. Both first and last draw lead, however, did not become normally distributed even after

log- or inverse (1/Y) transformations. Thus, to keep consistency with analytical methods, we used non-

parametric analyses throughout this analysis. WLL was also analyzed as a dichotomous outcome, and samples were classified by whether lead concentrations were detectable or exceeded the higher level quantiles of the total lead distribution (i.e. the 67th or 75th percentile values). Non-parametric analyses of WLL used the Wilcoxon Signed Rank tests, and county-specific differences of lead levels and corrosivity indices were analyzed using Mann-Whitney U tests to evaluate geographic and prospective geological phenomena. Housing stock differences across counties were also analyzed using t-tests. From there, we conducted the following bivariate analyses, detailed in TABLE VIII.

TABLE VIII- SUMMARY OF BIVARIATE ANALYSES

	Corrosivity	WLL

Age of Housing	1) Housing and Corrosivity: Continuous
	    a. Spearman's Rank correlation
	2) Housing: Dichotomous (Pre/post 1986) Corrosivity: Continuous
	    a. Mann-Whitney U test
	    b. t-approximated p-values
	3) Housing: Dichotomous (Pre/post 1986) Corrosivity: Categorical (empirical thresholds
	    a. Fisher's Exact Test
		1) Housing and WLL: Continuous
		    a. Spearman's Rank correlation
		2) Housing: Dichotomous (Pre/post 1986) WLL: Continuous
		    a. Mann-Whitney U test
		    b. t-approximated p-values
		3) Housing: Dichotomous (Pre/post 1986) WLL: Dichotomous (Detection limit status, 67th percentile status)
		    a. Fisher's Exact Test

WLL	1) Corrosivity and WLL: Continuous
	    a. Spearman's Rank correlation
	2) Corrosivity: Continuous WLL: Dichotomous (Detection limit status)
	    a. Mann-Whitney U test
	    b. t-approximated p-values
	3) Corrosivity: Categorical (Empirical thresholds) WLL: (Detection limit status, 67th percentile status)
	    a. Fisher's Exact Test	

Key to these analyses were the establishment of differences between WLLs before and after 1986. After we had found the housing cutpoints and had fully evaluated lead-corrosivity and corrosivity-housing relationships, we looked specifically at homes constructed before our housing cutpoint of interest (1986). We used similar methods as described above in TABLE VIII.

1) Corrosivity and WLL: Continuous

    a. Test: Spearman's Rank correlation

    b. Rationale: Evaluate the presence of linear or monotonic relationships between corrosivity and lead

2) Corrosivity: Continuous

    WLL: Dichotomous (Detection limit status, 67th percentile status)

    a. Test: Mann-Whitney U test; t-approximated p-values

    b. Rationale: to understand if corrosivity is higher in detectable lead samples and samples exceeding the 67th percentile

3) Corrosivity: Categorical (Empirical thresholds)

    WLL: (Detection limit status, 67th percentile status)

    a. Test: Fisher's Exact Test

    b. Rationale: Analyze discrete interactions between corrosivity and lead.

In addition to the bivariate comparisons depicted above, we also conducted a set of sensitivity analyses. We looked at both corrosivity variables and treated them by their empirical thresholds or by quantiles (specifically tertiles, quartiles and quintiles). We then looked at the association of each categorization of corrosivity with lead classifications by detectability and exceedance of percentiles (60th, 67th, 80th) to ensure the robustness of our effects, and to inform our logistic regression modelling.

Finally, we conducted exploratory multivariate logistic regression modelling to predict lead detection as our dichotomous outcome. *A priori*, we expected that increases in corrosivity would result in increases in water lead, but we did not have any assumptions regarding the strength or shape of this relationship. Thus, we used a few transformations, categorizations, and dichotomizations of our corrosivity variable (Larson Skold Index, in this case). Specifically, we used:

- Unaltered LSI, as a continuous variable

- Log-transformed LSI

- Quadratic (LSI^2, LSI)

- Various quantile comparisons (quartiles and quintiles) of LSI

- Dichotomized by empirical threshold (Low or Moderate/High) of LSI

- Dichotomized by the 75th percentile of LSI (above or below)

We also looked at age of housing as both a continuous predictor and dichotomized by pre/post 1986. From these results, we then modeled lead detection with both corrosivity and housing age cutpoints with and without interaction terms.

We took the housing age and corrosivity cutpoints yielding significant associations and used them as a "screening tool" for predicting lead detection within a given sample. We subsequently evaluated the positive predictive value (PPV) and the negative predictive value (NPV), as well as the sensitivity and specificity of our method.

<div align="center">

V.   RESULTS

</div>

A. <u>**Univariate Analyses**</u>

1. <u>**Lead**</u>

TABLE *IX* summarizes results of water lead levels. In a total of 97 samples, we had detectable

levels of lead in our first draw total lead (n=46, 47.4%) and our last draw last draw total lead samples

(n=18, 18.56%). For first draw lead samples with detectable lead, the mean was relatively low (3.16

µg/L, SE=0.96) in relation to the 15µg/L cutoff value under the Lead and Copper Rule. The median was

the same as the limit of detection (0.54 µg/L). Only samples exceeding the action level (n=3 or 3.1% of

the total) were further evaluated specifically for dissolved lead. None of the geochemical parameters

were distributed normally based on the Shapiro-Wilk test. The distribution of total water lead by

quantile (TABLE *X*) further illustrates the right skewness of the data. For the first draw samples,

detectable levels of lead were observed at the 67th percentile (1.57 µg/L), the 75th (2.16 µg/L) and the

80th (2.95 µg/L), while under detection limit values were observed at even the 80th percentile among the

last draw samples.

More samples came from Kane than Jackson (Kane: n=59, Jackson: n=38). In both counties,

detectable lead samples comprised a greater proportion of first draw (Jackson: 52.6%, Kane: 44.1%) and

last draw (Jackson 29.0%, Kane: 11.9%) total lead samples. First draw total lead samples from Jackson

County had a higher median and mean than those from Kane County (Median: Jackson: 0.85, Kane: 0.54;

Mean: Jackson=4.28, Kane=2.43). Last draw sample means were also higher for Jackson (0.88 vs 0.54),

but medians were both at the limit of quantification. The three samples with total lead leads above 15

µg/L (two from Jackson, one from Kane County) were mainly particulate lead. The first draw samples in

both Jackson and Kane County were similar to each other, and any differences were not statistically

significant **(**Mann-Whitney, p=0.60), but there was a statistically significant difference for last draw lead

samples by county (Mann-Whitney, p=0.03). This did not affect our analytical choices, however, since

first draw lead was the dominant source of elevated lead measurements for the purposes of our analysis.

The median and mean levels of lead for first draw samples were greater than last draw samples in the aggregate and by county. Paired Wilcoxon signed-rank tests found statistically significant differences between first and last draw lead when both counties were evaluated together ($p<0.0001$) and separately (Jackson: $p<0.0001$, Kane: $p<0.0001$).

2. Geochemical Parameters and Corrosivity

Both corrosivity metrics were derived from the same set of geochemical parameters, depicted in the aggregate and by county (TABLE *XI*). Only sulfate as a parameter was measured as undetectable (n=75, 75.3%), with most of the detected samples found in Jackson County (Jackson: n=36, 94.7%, Kane: n=37, 62.7%). Sulfate mean and median concentrations were also higher in Jackson (Mean- Jackson: 58.64, Kane: 29.25; Median- Jackson: 37.30, Kane: 17.4), while chloride levels were larger in Kane County (Mean- Kane: 80.66, Jackson: 49.55; Median- Kane: 36.8, Jackson: 8.90). We found statistically significant differences of chloride (Mann-Whitney U: $p=0.01$), sulfate (Mann-Whitney U: $p=0.003$), and alkalinity (Mann-Whitney U, $p=0.0002$) by county.

TABLE *XII* described our corrosivity metrics in the aggregate and by county. Neither of these indices were normally distributed in the aggregate or by county. Larson Skold Index distributions were comparable between individual counties and between counties and the aggregate across the distribution. They did not have any statistically significant variation by county (Mann-Whitney U: $p=0.54$). CSMR values are higher in Kane County (Mean- Kane: 64.89, Jackson: 2.73; Median- Kane: 4.91, Jackson: 2.10). Differences are orders of magnitude larger between Kane and Jackson County at higher quantiles of CSMR. The difference in CSMR distribution by county was statistically significant (Mann-Whitney U: $p<0.0001$).

TABLE IX- SUMMARY OF WATER LEAD DISTRIBUTION AND FRACTIONATION IN AGGREGATE AND BY COUNTY

	Variable	N[a]	# Detect.	Mean (µg/L)	Median (µg/L)	Max. (µg/L)	Std. Error (µg/L)	Shapiro-Wilk Test of Normality[c]
Both Counties	Tot. Pb- 1st Draw	97	46 (47.4%)	3.16	0.54	76.2	0.96	p<0.0001*
	Tot. Pb- Last Draw	97	18 (18.6%)	0.74	0.54	3.93	0.06	p<0.0001*
	Diss. Pb- 1st Draw [b]	3	2 (66.7%)	1.86	0.54	4.49	1.32	--
	Diss. Pb- Last Draw[b]	0	--	--	--	--	--	--
	Particul. Pb- First Draw [b]	3	3 (100%)	49.41	46.46	71.71	12.11	--
	Particul. Pb- Last Draw[b]	0	--	--	--	--	--	--
Jackson	Tot. Pb- 1st Draw	38	20 (52.6%)	4.28	0.85	76.2	2.1	p<0.0001*
	Tot. Pb- Last Draw	38	11 (29.0%)	0.88	0.54	3.93	0.12	p<0.0001*
	Diss. Pb- 1st Draw [b]	2	1 (50%)	2.52	2.52	4.49	1.98	--
	Diss. Pb- Last Draw [b]	0	--	--	--	--	--	--
	Particul. Pb- 1st Draw [b]	2	--	50.89	50.89	71.71	20.83	--
	Particul. Pb- Last Draw [b]	0	--	--	--	--	--	--
Kane	Tot. Pb- 1st Draw	59	26 (44.1%)	2.43	0.54	47	0.81	p<0.0001*
	Tot. Pb- Last Draw	59	7 (11.9%)	0.65	0.54	3.37	0.05	p<0.0001*
	Diss. Pb- 1st Draw [b]	1	0 (0.00%)	0.54	0.54	0.54	--	--
	Diss. Pb- Last Draw [b]	0	--	--	--	--	--	--
	Particul. Pb- 1st Draw [b]	1	--	46.46	46.46	46.46	--	--
	Particul. Pb- Last Draw [b]	0	--	--	--	--	--	--

[a] 1 sample has survey data and is captured in housing age analyses, but does not currently have lab data yet

[b] Dissolved lead data were only evaluated for samples above the EPA Action Level of 15 µg/L. Particulate lead was calculated by (Total Lead)-(Dissolved Lead)

[c] For Shapiro-Wilk Tests, p<0.05 means that the data are not normally distributed

*Significant at α=0.05

TABLE X- TOTAL LEAD QUANTILE DISTRIBUTION

Variable	N[a]	33rd	50th	67th	75th	80th
Total Lead- First Draw (µg/L)	97	0.54[b]	0.54	1.57	2.16	2.95
Total Lead- Last Draw (µg/L)	97	0.54	0.54	0.54	0.54	0.54

[a] 1 sample has survey data and is captured in housing age analyses, but does not currently have lab data yet

[b] 0.54 is the imputed value of the limit of detection by calculating $\frac{Sample\ QuantificationLimit}{\sqrt{2}}$

When we looked at corrosivity by established empirical thresholds for the Larson Skold Index (TABLE *XIII*), most samples had low corrosivity in the aggregate and by county.   A greater percentage of samples in Jackson County were potentially corrosive (21.2%) when compared to Kane County (13.6%). Because there were a limited number of samples labelled as "Moderate" and "High", we grouped these both together for the sake of statistical power.

Samples labelled by CSMR thresholds, by contrast, were mainly in the middle category of "Significant Concern of Corrosion" (81.6%) with no samples classified as "Serious Concern of Corrosion". Both Jackson and Kane County followed the same trend, with quite divergent distributions. There were drastically more Jackson County samples that reflected "No Concern" (Jackson: 34.2%, Kane: 6.9%), while more Kane County samples could be labelled as "Significant Concern" (Kane: 93.2%, Jackson: 65.8%). Because "Serious Concern of Corrosion" was not populated, we combined it with the Serious Concern category for future statistical analyses for more statistical power.

TABLE XI—SUMMARY OF GEOCHEMICAL PARAMETERS IN AGGREGATE AND BY COUNTY

	Variable	N[a]	# Detect.[b]	Mean (mg/L)	Std Error (mg/L)	Median (mg/L)	Max. (mg/L)	Shapiro-Wilk Test of Normality[c]
Both Counties	Alkalinity (CaCO3)	97	97 (100%)	338	10.10	344	725.00	p=0.003*
	Chloride	97	97 (100%)	68.47	13.17	25.10	1022.00	p<0.0001*
	Sulfate	97	75 (75.3%)	40.76	5.39	29.30	319.00	p<0.0001*
Jackson	Alkalinity (CaCO3)	38	38 (100%)	298.82	20.78	305.50	725.00	p=0.0396*
	Chloride	38	38 (100%)	49.55	26.85	8.90	1022.00	p<0.0001*
	Sulfate	38	36 (94.7%)	58.64	10.92	37.30	319.00	p<0.0001*
Kane	Alkalinity (CaCO3)	59	59 (100%)	364.80	8.41	358.00	582.00	p=0.0231*
	Chloride	59	59 (100%)	80.66	13.02	36.80	515.00	p<0.0001*
	Sulfate	59	37 (62.7%)	29.25	4.92	17.40	192.00	p<0.0001*

[a] 1 sample has survey data and is captured in housing age analyses, but does not currently have lab data yet

[b] Detection Limits: Sulfate: 0.21 mg/L, Chloride: 0.16 mg/L, Alkalinity: 4 mg/L

[c] For Shapiro-Wilk Tests, p<0.05 means that the data are not normally distributed

* Significant at $\alpha=0.05$

3. Age of Housing

The majority of housing (TABLE *XIV*) in the aggregate and by county were built pre-1986.

Proportionally, Kane County had slightly more pre-1986 housing stock (69.8%) than Jackson (60.0%).

Distributions by county were not found to be significantly different (p=0.94).

TABLE XII- SUMMARY OF CORROSIVITY METRICS IN THE AGGREGATE AND BY COUNTY

County	Variable	N[a]	Mean	Std Error	25th	50th (Med.)	75th	100th Max.	Shapiro-Wilk Test of Normality[b]
Both Counties	Larson Skold Index	97	0.41	0.05	0.07	0.23	0.59	2.24	p<0.0001*
	Chloride Sulfate Mass Ratio	97	40.5	14.47	0.36	2.10	7.61	1090.96	p<0.0001*
Jackson	Larson Skold Index	38	0.45	0.09	0.12	0.22	0.56	2.18	p<0.0001*
	Chloride Sulfate Mass Ratio	38	2.73	1.54	0.18	0.32	0.81	56.16	p<0.0001*
Kane	Larson Skold Index	59	0.39	0.06	0.06	0.26	0.63	2.24	p<0.0001*
	Chloride Sulfate Mass Ratio	59	64.82	23.29	1.98	4.91	24.45	1090.96	p<0.0001*

[a] 1 sample has survey data and is captured in housing age analyses, but does not currently have lab data yet

[b] For Shapiro-Wilk Tests, p<0.05 means that the data are not normally distributed

*Significant at α=0.05

B. Bivariate Analyses

1. Lead and Housing Age

Analysis of housing age as a continuous variable with total first draw indicated that there was a weakly negative monotonic relationship that approached significance (Spearman's ρ= -0.16, p=0.13); thus our treatment of housing age was predominantly categorical. While our *a priori* assumption was to use 1986 as a housing age cut off, we conducted a series of sensitivity analyses to better understand how dichotomization would affect the relationship between lead and housing (TABLE *XV*). The two cutpoints were lead levels were significantly different were around the median housing age (1982) and from our a priori threshold (1986). In both cases, there was a significant difference in the median lead levels which roughly corresponded to detection and non-detection (ND) in 1982 (Median: pre=1.11, post=0.54 (ND)) and 1986 (Median: pre=1.11, post=0.54 (ND)). It is important to note that the differences in the means before and after 1982 (Pre 1982: 3.43, Post 1982: 3.20) were consistent with

the differences in medians, while this did not hold to be true for average lead levels in homes

constructed pre and post 1986 (Pre 1986: 3.28, Post 1986: 3.43).

TABLE XIII- DISTRIBUTION OF CORROSIVITY VALUES BY EMPRICAL THRESHOLDS IN THE AGGREGATE AND BY COUNTY

Definition[1]	Empirical Threshold[2,3]	Both Counties (n=97)	Jackson (n=38)	Kane (n=59)
Chloride and sulfate concentrations will not affect natural scale formation (Low)	(LSI<0.8)	81 (82.7%)	30 (79.0%)	51 (86.4%)
Chloride and sulfate concentrations may affect natural scale formation, possibly causing corrosion (Moderate)	(0.8<LSI<1.2)	9 (9.2%)	3 (7.9%)	6 (10.2%)
High corrosion rates are anticipated (High)	(LSI>1.2)	7 (7.1%)	5 (13.2%)	2 (3.4%)
No corrosive concern	(CSMR <0.2)	17 (17.4%)	13 (34.2%)	4 (6.8%)
Significant concern of corrosion	(0.2<CSMR<0.5) or (CSMR >0.5 and Alkalinity >50 mg/L)	80 (81.6%)	25 (65.8%)	55 (93.2%)
Serious concern of corrosion	(CSMR >0.5 and Alkalinity <50 mg/L)	0 (0.0%)	0 (0.0%)	0 (0.0%)

Notes:

[1] Definitions as provided by Leitz & Guerra (2013) and Nguyen, Stone, and Edwards (2011)

[2] LSI thresholds describe the corrosivity of water towards mild steel, and were developed using Great Lakes water (Larson & Skold, 1958)

[3] CSMR thresholds are specifically for situations where there is galvanic corrosion-inducing settings (i.e. lead solder connected to copper piping)

TABLE XIV- DISTRIBUTION OF HOUSING STOCK AGE BY COUNTY

	N[1,2]	Mode	Built Pre-1986	Min. (0th)	25th	Med. (50th)	75th	Max. (100th)
All Counties	89	1978	58 (65.2%)	1850	1968	1978	1993	2013
Jackson	35	1977	21 (60.0%)	1880	1969	1984	1994	2013
Kane	53	1978	37 (69.8%)	1850	1968	1978	1993	2005

[1] One of these data points does not have lead data associated with it
[2] About 9 samples with lead results are still awaiting surveys or have not yet been processed. Distribution: Jackson (3) Kane (6)

Housing construction age status before and after 1986 was significantly associated with detection status (TABLE *XVI*). Greater proportions of post-1986 samples were below detection limit (n=23, 74.2%), while a larger proportion of pre-1986 samples had detectable lead (n=34, 58.6%) than houses constructed post-1986 (n=8, 25.8%). When evaluating the relationship of the upper tertile of lead to housing age, the majority of pre- and post-1986 homes by proportion were below the upper tertile of lead, with post-1986 homes comprising a larger proportion (Post 1986: n=34, 58.6%, Pre 1986: n=24, 77.4%). Greater proportions of pre-1986 homes had samples in the upper tertile of lead when compared post-1986 homes. However, no significant association was observed.

TABLE XV- EVALUATION OF HOUSING STOCK AGE DICHOTOMIZATION ON FIRST DRAW WATER LEAD LEVELS

Housing Age Cutoff	Construction Status Before/ After Cutoff	N	Mean	Std Error	Med.	Max.	Mann-Whitney U Test
1952 (10th percentile)	Before	11	1.57	0.44	1.07	4.38	
	After	78	3.58	1.2	0.54	76.2	p=0.68
1968 (25th percentile)	Before	24	1.72	0.37	0.88	7.25	
	After	65	3.93	1.44	0.54	76.2	p=0.77
1972 (33rd percentile)	Before	32	1.85	0.37	0.88	8.64	
	After	57	4.18	1.63	0.54	76.2	p=0.58
1982 (Median)	Before	51	3.43	1.51	1.11	76.2	
	After	38	3.2	1.43	0.54	47	p=0.02*
1986 (Regulatory Change)	Before	57	3.28	1.33	1.11	76.2	
	After	31	3.43	1.74	0.54	47	p=0.01*

*Significant at α=0.05

2. Corrosivity and Housing Age

The evaluation of corrosivity and housing was done to determine whether water corrosivity was associated with housing age, or if housing condition may confound corrosivity's function as an independent variable. The relationship between housing age and Larson Skold (Spearman's ρ=-0.02, p=0.86), and housing age with CSMR (Spearman's ρ=-0.02, p=0.88) did not show a significant or monotonic relationship.

The distribution of Larson Skold values by housing age (

TABLE *XVII*) showed no statistically significant differences between pre and post-1986 homes. Likewise, for CSMR there were no statistically significant differences between pre and post-1986 housing.

Corrosivity indices by pre/post-1986 classifications are described in

TABLE XVIII.   There was no statistically significant association between housing status and the Larson Skold empirical thresholds or the CSMR category. Of note is a lack of correspondence between corrosivity category based on the two measurement methods: only about 20% of samples were in the low corrosivity category based on CSMR data, while about 85% of samples were in the low corrosivity category based on the Larson Skold Index.

TABLE XVI- ASSOCIATION OF DETECTION AND UPPER TERTILE STATUS OF FIRST DRAW LEAD BY HOUSING AGE

Lead Threshold[1,2]	Pre-1986 Construction (n=58)	Post-1986 Construction (n=31)	Status Unknown (n=9)	Fischer's Exact Test
Detection Status[3]				
Below Detection Limit (n=51)	23 (39.7%)	23 (74.2%)	5 (55.6%)	
Above Detection Limit (n=46)	34 (58.6%)	8 (25.8%)	4 (44.4%)	p=0.01*
Missing Data	1 (1.7%)	0 (0.0%)	0 (0.0%)	
Above/Below 67th Percentile				
Below 67th Percentile (n=65)	34 (58.6%)	24 (77.4%)	7 (77.8%)	
Above 67th Percentile (n=32)	23 (39.7%)	7 (22.6%)	2 (22.2%)	p=0.34
Missing Data	1 (1.72%)	0 (0.0%)	0 (0.0%)	

*Significant at α=0.05
[1] One of these data points does not have lead data associated with it.
[2] About 9 samples with lead results are still awaiting surveys or have not yet been processed
[3] Detection limit is 0.76 µg/L, 67th percentile of lead is 1.57 µg/L

TABLE XVII- CORROSIVITY METRIC DISTRIBUTIONS BY HOUSING AGE (PRE/POST-1986)

Variable[1,2]	Age of Housing	N	Mean	Std Error	25th	Med. (50th)	75th	Max. (100th)	Mann-Whitney U
Larson Skold	Pre-86 House	57	0.42	0.06	0.09	0.22	0.63	1.59	
	Post-86 House	31	0.35	0.08	0.05	0.23	0.41	2.18	p=0.28
CSMR	Pre-86 House	57	46.12	22.5	0.42	1.98	6.6	1090.96	
	Post-86 House	31	28.4	16.6	0.22	2.3	7.8	497.67	p=0.79

[1] One of these data points does not have lead data associated with it

[2] About 9 samples with lead results are still awaiting surveys or have not yet been processed

TABLE XVIII- CLASSIFICATION OF SAMPLES BY CORROSIVITY EMPIRICAL THRESHOLDS AND HOUSING STATUS

Corrosion Indices and Thresholds[1]	Pre-86[2] (N=58)	Post-86 (N=31)	Fisher's Exact
Larson Skold			
Low	46 (79.3%)	28 (90.3%)	p=0.61
Moderate/High[3]	11 (19.0%)	3 (9.7%)	
Moderate	6 (10.3%)	2 (6.5%)	Levels included for descriptive purposes
High	5 (8.6%)	1 (3.2%)	
CSMR			
No Concern	10 (17.2%)	7 (22.6%)	p=0.50
Significant/Serious Concern	47 (81.0%)	24 (77.4%)	

[1] 9 samples with lead results are still awaiting surveys or have not yet been processed.
[2] One of these data points does not have lead data associated with it
[3] Fisher's Exact Tests carried out for Larson Skold using Low and Moderate/High as comparisons.

3. Corrosivity and Lead

Correlational analysis yielded a weak positive relationship between increasing Larson Skold and first draw lead that was of borderline statistical significance (Spearman's $\rho=0.17$, $p=0.11$), and no association between CSMR and first draw lead (Spearman's $\rho=-0.13$, $p=0.21$).

We found that median Larson Skold Index values were higher for detectable lead samples (Above Detection: 0.28, Below Detection: 0.16), but it was not a significant difference (TABLE *XIX*). By contrast, CSMR median values for detectable lead samples were higher than non-detectable, but this difference was also insignificant.

We also looked at the associations of corrosivity expressed by its empirical thresholds in association with detectable lead and upper tertile lead (67th percentile) status (TABLE *XX*). Looking at the Larson Skold Index values, the majority of samples regardless of detection status were considered to have Low corrosivity. However, twice the percentage of above detection samples were associated with Moderate/High than those below the detection limit (Above Detection: 21.7%, Below Detection: 11.8%). This association was statistically significant. We found similar results for CSMR empirical categorizations.   The above detection limit samples comprised a larger percentage of the No Concern category, while slightly more below detection limit samples were considered to be Significant/Serious Concern. This association was also statistically significant.

For upper tertile lead status, we found that the proportions of Low corrosivity samples below and above the 67th percentile were similar. However, the proportion of Moderate/High corrosivity samples above the upper tertile were 1.5 times higher (Above 67: n=7, 21.9%; Below 67: n=9, 13.9%).

This association was statistically significant. For CSMR, we found that there were slightly more No

Concern samples below the 67th percentile than above it. Among the Significant/Serious category, a

slightly higher proportion had above 67th percentile lead. Both differences in No Concern and

Significant/Serious between upper tertile lead status varied by around 3 percentage points. This

association was also statistically significant.

TABLE XIX- AVERAGE AND MEDIAN CORROSIVITY OF FIRST DRAW LEAD SAMPLES BY LEAD DETECTION STATUS

Corrosivity Measure	Lead Threshold[1]	N[2]	Group Means	Group Medians	Mann-Whitney U[3]
Larson Skold Index	Below Detection Limit	51	0.37	0.16	p=0.13
	Above Detection Limit	46	0.47	0.28	
Chloride Sulfate Mass Ratio	Below Detection Limit	51	19.72	3.28	p=0.19
	Above Detection Limit	46	63.53	1.55	

[1] Detection limit is 0.76 µg/L.
[2] 1 sample missing lab data, but has survey information
[3] Used t-approximation for p-values

C. Corrosivity and Lead Association in Pre-1986 Homes

Figure 3 depicts the overall relation relationship of water lead, housing age status, and the

Larson Skold thresholds, indicating differences in means between the Low and Moderate/High

categories of pre-1986 lead. Water samples from post-1986 homes show that the largest average and

median water lead had Moderate corrosivity. Looking at trends for CSMR (Figure 4), we do not see a

similar threshold effect at the different levels of CSMR. Thus, we decided to more specifically look at

corrosivity within the pre-1986 homes, with a focus on the Larson Skold Index.

1.   **Descriptive Analysis of Pre-1986 Water and Housing Variables**

TABLE *XXI* is a summary of all parameters for pre-1986 first draw lead samples. The mean of

first draw (3.28 µg/L) lead was larger than the median (1.11 µg/L), and was also larger than the upper

tertile of the entire data set (1.57 µg/L). The majority of samples were detectable, and almost 40% were

above the 67th percentile. The IQR of lead was also larger than that of the complete dataset (Pre-86:

2.40, All Houses: 1.62), indicating that there was a wider spread of the data. In terms of housing age,

the mode was still the same as that of the aggregate.

Parameters of Larson Skold indicated that the mean and the median were both well within the

Low empirical threshold, and that the vast majority of samples were classified as Low (n=46, 80.70%).

The IQR of the Larson Skold Index values indicated that its distribution variability was almost identical to

the broader dataset (Pre-86: 0.54, All Houses: 0.52). The CSMR parameters were consistent with those

of the broader dataset, with a larger mean than median, indicating a skewed distribution. The majority

of samples were also classified as Significant Concern (n= 47 82.5%). However, the IQR was lower in the

pre-1986 homes than the broader dataset (Pre-86: 6.18, All Houses: 7.25), indicating a tighter

distribution.

2.   **Treatment of Corrosivity as Continuous**

Using samples from pre-1986 homes, we evaluated if linear relationships might exist between

corrosivity and first draw lead (Figure 5). Correlations of Larson Skold Index values and first draw lead

yielded an insignificant, weak association (Spearman's ρ= 0.20 p=0.15). CSMR was not correlated to first

draw lead (Spearman's ρ= -0.04, p=0.80).

TABLE XX- ASSOCIATION OF FIRST DRAW LEAD DETECTION AND CORROSIVITY EMPIRICAL THRESHOLD

Corrosion Indices and Thresholds	Below Detection Limit[1] (N=51)	Above Detection Limit (n=46)	Fischer's Exact P-value	Below 67th Percentile (N=65)	Above 67th Percentile (n=32)	Fischer's Exact P-value
Larson Skold Index[3]						
Low	45 (88.2%)	36 (78.3%)	p=0.01*	56 (86.2%)	25 (78.1%)	p=0.001*
Moderate/High[5]	6 (11.8%)	10 (21.7%)		9 (13.9%)	7 (21.9%)	
Moderate	4 (7.8%)	5 (10.9%)	Levels are described for descriptive purposes	5 (7.7%)	4 (12.5%)	Levels are described for descriptive purposes
High	2 (3.9%)	5 (10.9%)		4 (6.2%)	3 (9.4%)	
CSMR[4]						
No Concern	8 (15.7%)	9 (19.6%)	p=0.02*	12 (18.5%)	5 (15.6%)	p=0.02*
Significant/Serious Concern	43 (84.3%)	37 (80.4%)		53 (81.5%)	27 (84.4%)	

*Significant at α=0.05

[1] Detection limit is 0.76 µg/L. The 67th percentile of lead is 1.57 µg/L.

[2] About 1 sample missing lab data, but has survey information

[3] Larson Skold: Low (LR < 0.8), Moderate: (0.8 < LR < 1.2), High: (LR > 1.2)

[4] CSMR: No Concern: (CSMR < 0.2), Significant Concern (0.2 < CSMR < 0.5) or (CSMR > 0.5 and Alkalinity > 50 mg/L), Serious Concern (CSMR > 0.5 and Alkalinity < 50 mg/L)

[5] Fisher's Exact Tests carried out for Larson Skold using Low and Moderate/High as comparisons.

Figure 3- First draw lead by Larson Skold Index thresholds and housing age status[1,2]
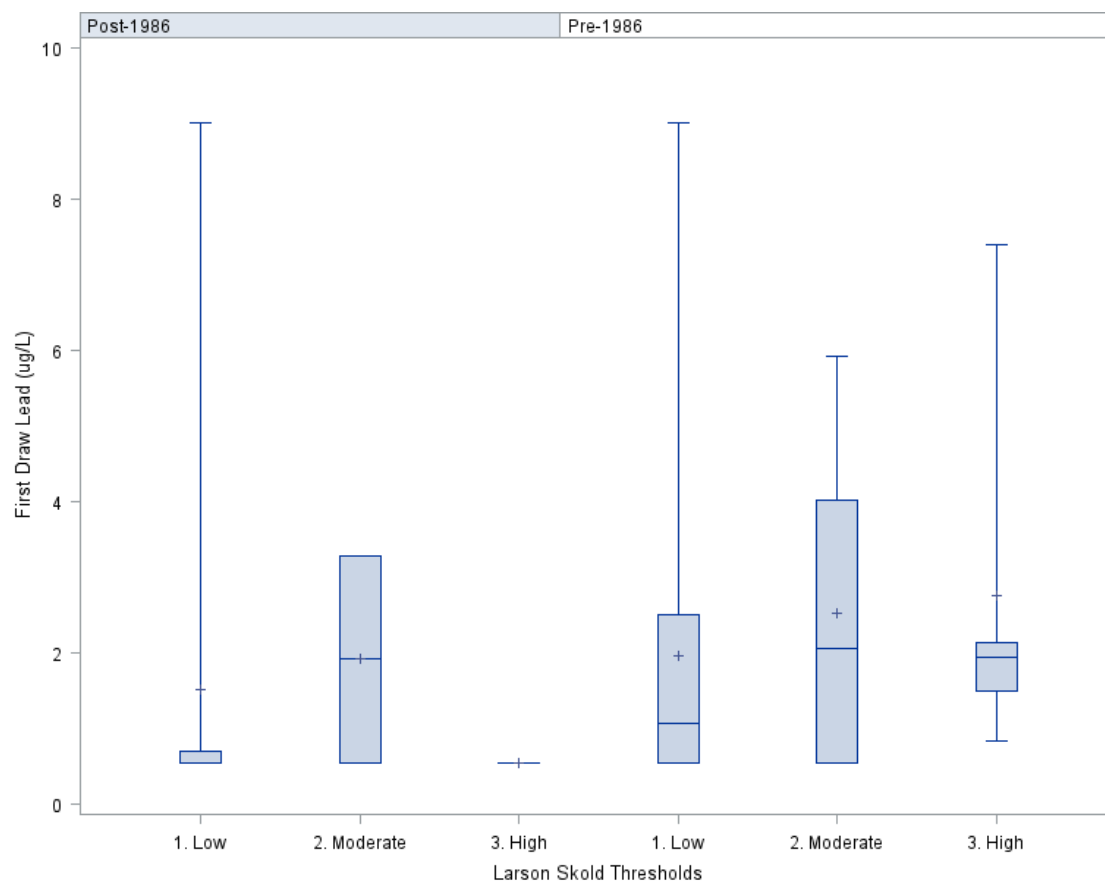
[1]Samples exceeding the EPA Action Level of 15 µg/L (n=3) were assigned a value of 9 for the purposes of making visualization of trends easier.
[2]Samples lacking corrosivity data associated with them were excluded(n=1).

Figure 4- First draw lead by chloride-sulfate mass ratio thresholds and housing age status[1,2]
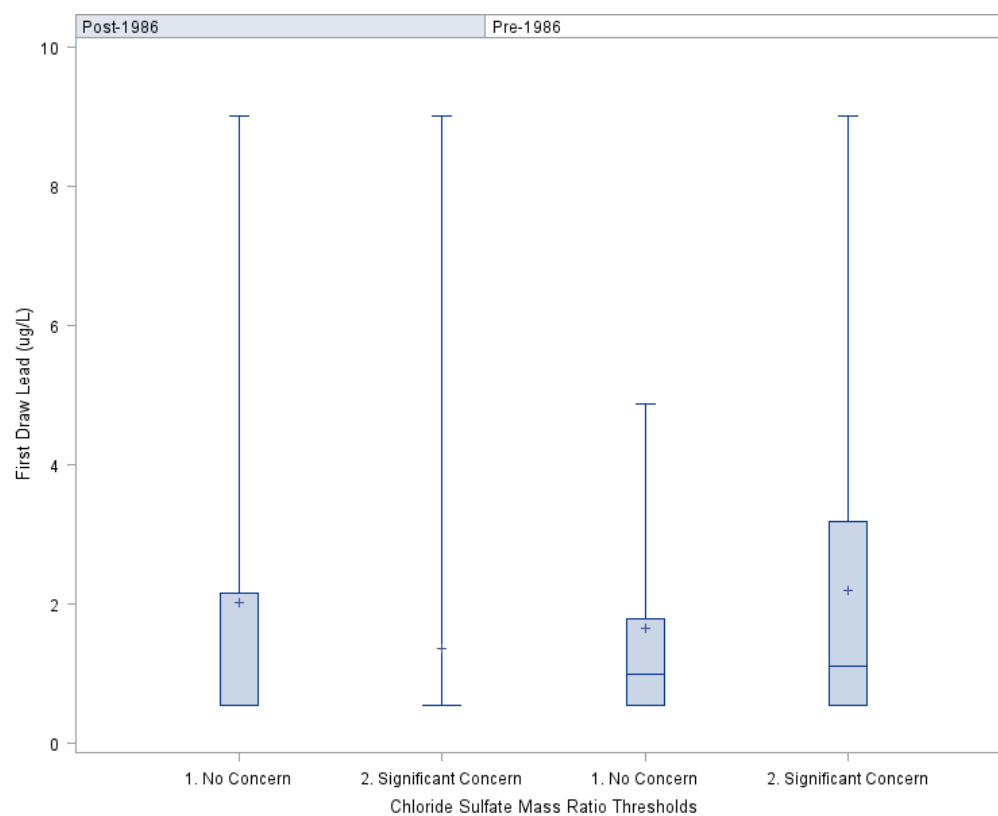
[1]Samples exceeding the EPA Action Level of 15 μg/L (n=3) were assigned a value of 9 for the purposes of making visualization of trends easier.
[2]Samples lacking corrosivity data associated with them were excluded(n=1).

TABLE XXI- SUMMARY OF ALL PRE- 1986 FIRST DRAW LEAD SAMPLES

Parameters	
First Draw Lead	n=57[1]
Mean	3.28
SE	1.33
Median	1.11
IQR	2.40
Above Detection Limit[2]	34 (59.7%)
Above 67 Percentile[3]	23 (40.4%)
Housing	n=58
Median Year	1970
Mode	1978
Corrosivity	
Larson Skold Index[4]	n=57*
Mean	0.43
SE	0.06
Median	0.22
IQR	0.54
LSI Empirical Thresholds	
Low	46 (80.7%)
Moderate	6 (10.5%)
High	5 (8.8%)
Chloride Sulfate Mass Ratio[5]	n=57*
Mean	46.12
SE	22.52
Median	1.98
IQR	6.18
CSMR Empirical Thresholds	
No Concern	10 (17.5%)
Significant Concern	47 (82.5%)
Serious Concern	0 (0.0%)

[1] 1 sample with survey data is missing corresponding lab data

[2] Detection limit of lead is 0.76 µg/L

[3] 67th percentile of lead is 1.57 µg/L

[4] Larson Skold: Low (LR < 0.8), Moderate: (0.8 < LR < 1.2), High: (LR > 1.2)

[5] CSMR: No Concern: (CSMR < 0.2), Significant Concern (0.2 < CSMR < 0.5) or (CSMR > 0.5 and Alkalinity > 50 mg/L), Serious Concern (CSMR > 0.5 and Alkalinity < 50 mg/L)

We also evaluated the differences between our corrosivity metrics as continuous by lead detection and 67th percentile status testing (TABLE *XXII*). No statistically significant differences in rank corrosivity values were noted between dichotomous lead categories (detection, 67th percentile) for either corrosivity metric. Differences in Larson Skold group medians by 67th percentile status were larger than the comparison with detection status, and the former's p-value approached marginal significance.

3. **Corrosivity by Empirical Thresholds**

Analysis of first draw lead concentrations by Larson empirical thresholds showed that median lead levels were higher in the Moderate/High category, (

TABLE XXIII) but overall showed no statistically significant differences. Comparison of median lead levels by the CSMR empirical thresholds showed lead was elevated in the Significant Concern category, but was also not significant.

We used these same corrosivity empirical thresholds and compared them with their lead detection status (TABLE *XXIV*). A greater percentage of Larson-Skold Low corrosivity values were below the lead detection limit. Additionally, we also found over triple the proportion of above detection limit samples had Moderate/High corrosivity when compared to those below detection (Above Detection: n=9, 26.5%; Below Detection: n=2, 8.7%). This association was statistically significant (p=0.01). For CSMR, we found similar proportions of No Concern corrosivity among samples that were both below and above detection. The same trend was also observed among samples labeled Significant Concern, with the majority of samples for both detection levels having Significant Concern (Below Detection: n=19, 82.6%, Above Detection: n=28, 82.4%. This association was statistically significant (p=0.04).

We also evaluated corrosivity thresholds to a sample's upper lead tertile status. For Larson Skold, we found that a greater percentage of samples below 67th percentile lead had Low corrosivity. About twice the proportion of samples that were above 67th percentile lead was labelled as Moderate/High corrosivity (Above 67th: n=6, 26.1%, Below 67th: n=5, 14.7%). This association was statistically significant (p=0.016). CSMR similarly had a greater percentage of below 67th percentile lead samples with No Concern corrosivity. We also found that a greater proportion of above 67th percentile lead samples had Significant corrosivity, but this was only a 7.5 percentage point difference (Above 67th: n=20, 87.0%; Below 67th: n=27, 79.4%). This association was also statistically significant (p=0.03).

D. Samples Above EPA Action Level (15 μg/L)

TABLE *XXV* contains all samples exceeding the EPA's action level set for community water systems. In this limited number of samples, 2 of 3 were constructed after 1986. The Larson Skold Index values are well within the Low threshold, while two of them have Significant CSMR values. The lead composition of these three samples is primarily particulate lead. Only one of them had detectable levels of dissolved lead, but even in this case, the sample was composed of 94% particulate lead.

E. Logistic Regression Modelling

TABLE XXVI and TABLE XXVII display the results of our single predictor exploratory logistic regression modelling. We started out with univariate modelling of lead detection with Larson Skold Index (LSI) and age of housing as predictors. Transformations applied to the LSI for these single predictor models include taking the natural log (ln(LSI), treating LSI as a quadratic (LSI^2 + LSI) and division of LSI into quantiles (quartiles and quintiles) with comparisons of the upper quantiles to the lowest quantile. We also used a few additional dichotomizations, specifically based on whether LSI exceeded or was below the 4th Quartile (0.59), and if the LSI value of a sample had Moderate and High Corrosivity or not (>0.8). For LSI, we found that about 4 of our 7 single-predictor models produced a significant odds ratio estimate. We found a statistically significant odds ratio between Quintile 3 and 1 (OR: 3.71, 95% CI:

1.06, 14.29), and between Quartile 4 and 1 (OR: 3.56, 95% CI: 1.13, 12.08). Additionally, we found that

our specifically dichotomized between below and above Quartile 4 was similarly significant. LSI in the

quadratic model was significant (OR: 10.22, 95% CI: 1.04, 122.67), but not LSI^2. We believe these

nonlinearities may have been better summarized in the quantile models.

Most of the parameter estimates were not significant. Based on LSI quartile-specific

associations with water lead detection, the parameter estimate for the comparison between Quartile 4

and Quartile 1 was statistically significant ($\beta_{LSI\,Q4,\,Q1}$= 1.27, p<0.05), as well as the parameter estimate

dichotomizing below and above 0.59 ($\beta_{LSI\,Q4}$= 1.05, p<0.05). The parameter estimates for the quadratic

model (LS^2 + LS) appeared to approach significance ($\beta_{LS}{}^2$: -1.14, p=0.10, β_{LS}: 2.32, p=0.052).

The odds ratio estimates of our multivariate modelling are available in
[a] Listwise deletion of 1 observation (n=97)
[b] Listwise deletion of 10 observations (n=88)
[c] Quintiles of Larson Skold Index are:
       Qunitile 1: 0.01-0.05 (20th percentile), Quintile 2: 0.06-0.16 (40th percentile), Quintile 3: 0.17-0.34 (60th percentile),
       Quintile 4: 0.34-0.74 (80th percentile), Quintile 5: >0.74
[d] Quartiles of Larson Skold Index are: Quartile 1: 0.01-0.07 (25th percentile), Quartile 2: 0.08-0.23 (50th percentile), Quartile 3:
0.24-0.59 (75th percentile), Quartile 4: >0.59
*Marginally significant at α=0.05 (Wald Chi Square)
**Significant at α=0.05

TABLE *XXVIII*, and our beta parameters are available in TABLE *XXIX*. We analyzed models with

and without an interaction term between corrosivity and pre-1986 housing construction status. Models

1 (LSI as continuous), 3 (natural log of LSI), and 5 (Dichotomization, if LSI values were considered

"Moderate" as a minimum) modelled lead detection with the assumption that there was no interaction

effect in the presence of an older, pre-1986 home. In all but two of these models (Model 2 and Model

4), the pre-1986 parameter was statistically significant. In all models, the odds of lead detection in the

presence of pre-1986 home was 4 times as likely when compared to newer homes after adjusting for

corrosivity. The three models without an interaction term did not show a significant association

between their respective form of the LSI with lead detection. When looking at models with an

interaction term (Models 2, 4, 6), there was also no significant interaction parameter or strata-specific

effects with the presence or absence of a pre-1986 house.

Figure 5- Relationship of Larson Skold (A) and CSMR (B) with first draw lead in pre-1986 homes[1,2]

Correlation of Larson Skold Corrosivity and First Draw Lead in Pre-1986 Homes

Observations	57
Spearman's Rho Correlation	0.19
p-Value	0.15

A

Correlation of CSMR Corrosivity and First Draw Lead in Pre-1986 Homes

Observations	57
Spearman's Rho Correlation	-0.04
p-Value	0.80

B
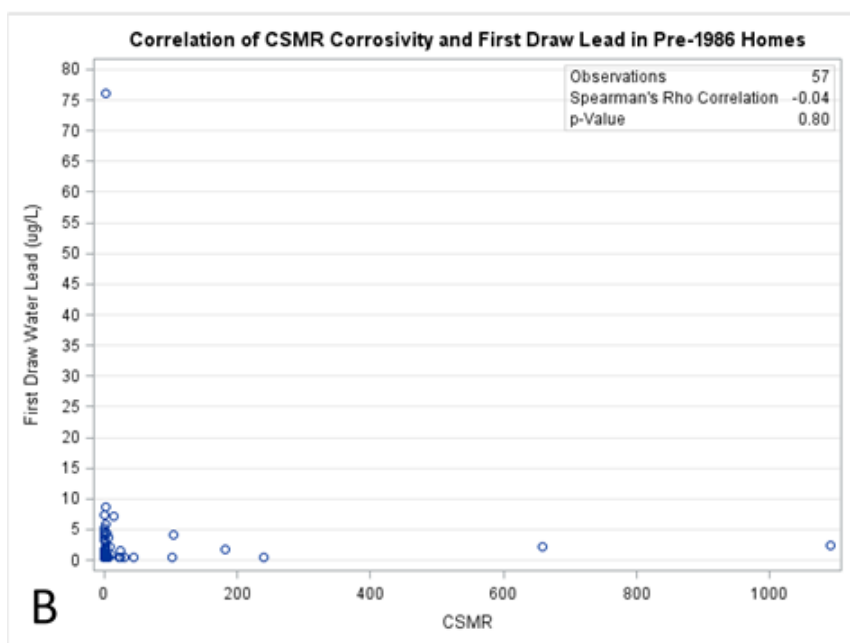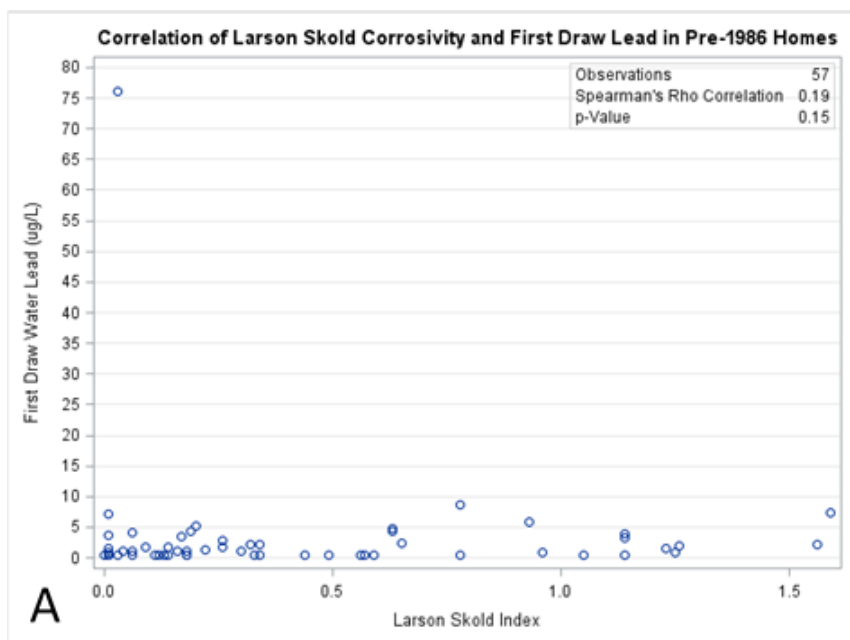
[1] About 1 sample with survey data is missing corresponding lab data
[2] $\alpha=0.05$

TABLE XXII- COMPARISON OF PRE-1986 CORROSIVITY METRICS BY FIRST DRAW LEAD DETECTION AND TERTILE OF DISTRIBUTION STATUS

Corrosivity Measure	Lead Threshold	N[1,2]	Group Means	Group Medians	Whitney-Mann U Test[3]
Larson Skold	Below Detection Limit[3]	23	0.32	0.18	p=0.21
	Above Detection Limit	34	0.49	0.26	
	Below 67th Percentile[4]	34	0.35	0.18	p=0.11
	Above 67th Percentile	23	0.54	0.54	
Chloride Sulfate Mass Ratio	Below Detection Limit	23	21.57	2.10	p=0.66
	Above Detection Limit	34	62.72	1.89	
	Below 67th Percentile	34	15.96	2.03	p=0.61
	Above 67th Percentile	23	90.7	1.93	

[1] 1 sample with survey data is missing corresponding lab data
[2] 9 samples with lead results are still awaiting surveys or have not yet been processed
[3] Used t-distribution for deriving p-value
[4] Detection limit of lead is 0.76 µg/L,
[5] 67th percentile of lead is 1.57 µg/L

TABLE XXIII- COMPARISON OF PRE-1986 FIRST DRAW LEAD BY CORROSIVITY EMPIRICAL THRESHOLDS CLASSIFICATION

Corrosivity Thresholds	N[1,2]	Group Means	Group Medians	Mann Whitney U
LR[4]	n=57			
Low	46	3.43	1.06	
Moderate/High[3]	11	2.63	1.95	p=0.19
Moderate	6	2.52	2.06	
High	5	2.76	1.95	
CSMR	n=57			
No Concern	10	1.66	1.00	
Significant/Serious Concern	47	3.62	1.11	p=0.70

[1] Used t-distribution for deriving p-value
[2] About 1 sample with survey data is missing corresponding lab data. About 9 samples with lead results are still awaiting surveys or have not yet been processed.
[3] Fisher's Exact Tests carried out for Larson Skold using Low and Moderate/High as comparisons. Moderate and High means are provided for descriptive purposes
[4] Larson Skold: Low (LR < 0.8), Moderate: (0.8 < LR < 1.2), High: (LR > 1.2)
[5] CSMR: No Concern: (CSMR < 0.2), Significant Concern (0.2 < CSMR < 0.5) or (CSMR > 0.5 and Alkalinity > 50 mg/L), Serious Concern (CSMR > 0.5 and Alkalinity < 50 mg/L)

TABLE XXIV- COMPARISON OF PRE-1986 CORROSIVITY THRESHOLDS BY FIRST DRAW LEAD DETECTION AND TERTILE OF DISTRIBUTION STATUS

Corrosivity Thresholds[1]	Below Detection Limit[2] (n=23)	Above Detection Limit (n=34)	Fisher's Exact P-value	Below 67th Percentile[3] (n=34)	Above 67th Percentile (n=23)	Fisher's Exact P-value
Larson Skold[4]						
Low	21 (91.3%)	25 (73.5%)	p=0.01*	29 (85.3%)	17 (73.9%)	p=0.02*
Moderate/High[5]	2 (8.7%)	9 (26.5%)		5 (14.7%)	6 (26.1%)	
Moderate	2 (8.7%)	4 (11.8%)	Levels are described for descriptive purposes	3 (8.8%)	3 (13.0%)	Levels are described for descriptive purposes
High	0 (0.00%)	5 (14.7%)		2 (5.9%)	3 (13.0%)	
CSMR[6]						
No Concern	4 (17.4%)	6 (17.7%)	p=0.04*	7 (17.7%)	3 (13.0%)	p=0.03*
Significant/Serious Concern	19 (82.6%)	28 (82.4%)		27 (79.4%)	20 (87.0%)	

*Significant at α=0.05

[1] 1 sample with survey data is missing corresponding lab data

[2] Detection limit of lead is 0.76 µg/L

[3] 67th percentile of lead is 1.57 µg/L

[4] Larson Skold: Low (LR < 0.8), Moderate: (0.8 < LR < 1.2), High: (LR > 1.2)

[5] Fisher's Exact Tests carried out for Larson Skold using Low and Moderate/High as comparisons.

[6] CSMR: No Concern: (CSMR < 0.2), Significant Concern (0.2 < CSMR < 0.5) or (CSMR > 0.5 and Alkalinity > 50 mg/L), Serious Concern (CSMR > 0.5 and Alkalinity < 50 mg/L)

TABLE XXV- ELEVATED FIRST DRAW LEAD SAMPLES ABOVE EPA ACTION LEVEL (15 µg/L)

County	Sample ID	Year of House Construction	Larson Skold Index	CSMR	Total[a] Lead (µg/L)	Particulate Lead (µg/L)	Dissolved Lead (µg/L)
Jackson	JA-21	1976	0.03 (Low)	2.17 (Signif.)	76.2	71.71	4.49
Jackson	JA-36	2011	0.23 (Low)	0.12 (None)	30.6	30.06	ND
Kane	KA-40	2004	0.01 (Low)	1.15 (Signif.)	47	46.46	ND

[a]First draw lead sample

TABLE XXVI- SUMMARY OF SINGLE-PREDICTOR LOGISTIC REGRESSION MODELS OF WATER LEAD DETECTION

Model#	Variables	Variable Type	Odds Ratio[e]
1[a]	LSI	Continuous	1.61 (0.68, 4.03)
2[a]	Ln(LSI)	Continuous	1.18 (0.90, 1.57)
3[a]	(LSI)^2	Continuous (Quadratic)	0.32 (0.07, 1.17)
	LSI		10.22 (1.04, 122.67)
4[a]	Quintiles (LSI)[c]	Ordinal	1.27 (0.95, 1.70)
		Quintile 2 vs 1	1.17 (0.31, 4.36)
		Quintile 3 vs 1	3.71 (1.06, 14.29)
		Quintile 4 vs 1	1.60 (0.44, 6.03)
		Quintile 5 vs 1	2.75 (0.78, 10.41)
5	Quartiles (LSI)[d]	Quartile 2 vs 1	1.64 (0.53, 5.21)
		Quartile 3 vs 1	1.14 (0.35, 3.73)
		Quartile 4 vs 1	3.56 (1.13, 12.08)
6[a]	LSI (as Moderate/High)	Dichotomous (yes, no)	2.08 (0.71, 6.64)
7[a]	Quartile 4 (LSI)	Dichotomous (yes, no)	2.87 (1.11, 7.88)
8[b]	Age of House	Continuous	0.98 (0.96, 1.00)
9[b]	House Older than 1986	Dichotomous (yes, no)	2.08 (1.68, 11.68)

[a] Listwise deletion of 1 observation (n=97)

[b] Listwise deletion of 10 observations (n=88)

[c] Quintiles of Larson Skold Index are:

Qunitile 1: 0.01-0.05 (20th percentile), Quintile 2: 0.06-0.16 (40th percentile), Quintile 3: 0.17-0.34 (60th percentile), Quintile 4: 0.34-0.74 (80th percentile), Quintile 5: >0.74

[d] Quartiles of Larson Skold Index are: Quartile 1: 0.01-0.07 (25th percentile), Quartile 2: 0.08-0.23 (50th percentile), Quartile 3: 0.24-0.59 (75th percentile), Quartile 4: >0.59

[e] 95% CI does not overlap with 1

*Marginally significant at α=0.05 (Wald Chi Square)

**Significant at α=0.05

TABLE XXVII- BETA PARAMETERS OF SINGLE-PREDICTOR LOGISTIC REGRESSION MODELS OF WATER LEAD DETECTION

Model#	Variables	Variable Type	β_1 (with CI)
1[a]	LSI	Continuous	0.47 (-0.40, 1.35)
2[a]	Ln(LSI)	Continuous	0.17 (-0.11, 0.44)
3[a]	$(LSI)^2$	Continuous (Quadratic)	-1.14 (-2.50, 0.23)
	LSI		2.32* (-0.02, 4.67)
4[a]	Quintiles (LSI)[c]	Ordinal	0.23 (-0.05, 0.52)
		Quintile 2 vs 1	0.15 (-1.15, 1.45)
		Quintile 3 vs 1	1.31 (0.02, 2.60)
		Quintile 4 vs 1	0.47 (-0.83, 1.77)
		Quintile 5 vs 1	1.01 (-0.27, 2.30)
5	Quartiles (LSI)[d]	Quartile 2 vs 1	0.50 (-0.64, 1.63)
		Quartile 3 vs 1	0.13 (-1.04 ,1.30)
		Quartile 4 vs 1	1.27** (0.09, 2.45)
6[a]	LSI (as Moderate/High)	Dichotomous (yes, no)	0.73 (-0.37, 1.84)
7[a]	Quartile 4 (LSI)	Dichotomous (yes, no)	1.05** (0.09, 2.02)
8[b]	Age of House	Continuous	-0.02* (-0.03, 0.002)
9[b]	House Older than 1986	Dichotomous (yes, no)	1.45** (0.48, 2.41)

[a] Listwise deletion of 1 observation (n=97)
[b] Listwise deletion of 10 observations (n=88)
[c] Quintiles of Larson Skold Index are:
    Qunitile 1: 0.01-0.05 (20th percentile), Quintile 2: 0.06-0.16 (40th percentile), Quintile 3: 0.17-0.34 (60th percentile), Quintile 4: 0.34-0.74 (80th percentile), Quintile 5: >0.74
[d] Quartiles of Larson Skold Index are: Quartile 1: 0.01-0.07 (25th percentile), Quartile 2: 0.08-0.23 (50th percentile), Quartile 3: 0.24-0.59 (75th percentile), Quartile 4: >0.59
*Marginally significant at α=0.05 (Wald Chi Square)
**Significant at α=0.05

TABLE XXVIII- MULTIVARIATE LOGISTIC REGRESSION OF WATER LEAD DETECTION, LARSON SKOLD INDEX, AND PRE-1986 HOUSING

Model[1]	Larson Skold Variable	OR (95% CI)[3]	
		Larson Skold	Pre-1986 Homes
1	Continuous, untransformed	2.15 (0.78, 6.50)	4.15 (1.61, 11.55)
2	Continuous, untransformed with interaction term	2.64 (0.69, 10.05)	
3	Ln (LSI)	1.19 (0.88, 1.63)	4.03 (1.72, 12.22)
4	Ln (LSI), with interaction term	1.13 (0.79, 1.61)	
5	Moderate/High (Dichotomous, yes/no)	2.89 (0.83, 11.87)	4.01 (1.56, 11.17)
6	Moderate/High, with interaction term	3.78 (0.73, 19.45)	
7	Quartile 4 (>0.59)[2] (Dichotomous, yes/no)	3.38 (1.15, 11.03)	3.99 (1.53-11.25)
8	Quartile 4, with interaction term	4.13* (1.02, 16.68)	

[1]Listwise deletion of 10 observations (n=88)

[2] Quartiles of Larson Skold Index are: Q1: 0.01-0.07 (25th percentile), Q2: 0.08-0.23 (50th percentile), Q3: 0.24-0.59 (75th percentile), Q4: >0.59

[3]95% CI does not overlap with 1

*Significant at α=0.05

TABLE XXIX- BETA PARAMETERS OF MULTIVARIATE LOGISTIC REGRESSION OF WATER LEAD DETECTION, LARSON SKOLD INDEX, AND PRE-1986 HOUSING

Model[a]	Larson Skold Variable	Beta Parameters (95% CI)		
		Larson Skold	Pre-1986 Homes	Interaction Term
1	Continuous, untransformed	0.77 (-0.27, 1.80)	1.42** (0.45, 2.40)	---
2	Continuous, untransformed with interaction term	0.42 (-1.31, 2.15)	1.21* (-0.06, 2.48)	0.55 (-1.63, 2.74)
3	Ln (LSI)	0.17 (-0.14, 0.48)	1.48** (0.51, 2.46)	---
4	Ln (LSI), with Interaction term	0.33 (-0.33, 0.98)	1.16 (-0.35, 2.66	-0.21 (-0.95, 0.54)
5	Moderate/High (Dichotomous, yes/no)	1.06 (-0.24, 2.36)	1.39** (0.41, 2.37)	---
6	Moderate/High, with interaction term	0.41 (-2.14, 2.95)	1.27** (0.24, 2.31)	0.92 (-2.11, 3.95)
7	Quartile 4 (>0.59)[b] (Dichotomous, yes/no)	1.22** (0.11, 2.33)	1.38** (0.39, 2.37)	---
8	Quartile 4, with interaction term	0.80 (-1.21 2.81)	1.25** (0.15, 2.35)	0.62 (-1.83, 3.07)

[a]Listwise deletion of 10 observation (n=88)
[b] Quartiles of Larson Skold Index are: Q1: 0.01-0.07 (25th percentile), Q2: 0.08-0.23 (50th percentile), Q3: 0.24-0.59 (75th percentile), Q4: >0.59
*Marginally significant at $\alpha=0.05$ (Wald Chi Square)
**Significant at $\alpha=0.05$

Model 7, by contrast, which modelled LSI in terms of whether a given sample's value exceeded the 75th percentile (0.59) with pre-1986 housing without an interaction parameter, exhibited a significant association of both corrosivity (OR=3.38, 95% CI: 1.15,11.03) and pre-1986 housing (OR=3.99, 95% CI: 1.53,11.25) with lead detection. The modelled interaction term of both of these parameters (Model 8) was not significant, but did yield a significant stratum specific effect (Pre-86 House*4th Quartile Larson Skold OR= 4.13, 95% CI: 1.02, 16.68).

F. Evaluation of Screening Decision Rules

Using the results of the multivariable modelling, we classified samples as originating from "High Risk Homes" if they had a Larson Skold Index value greater than 0.59 and originated from a home built

before 1986. TABLE *XXX* describes this classification scheme in relation to overall lead detection. We

found that our decision rule had high specificity (93.5%), but very low sensitivity (31.0%). It is important

to note that there were 9 samples that lacked housing level data and could not be classified for this

analysis, as well as 1 sample whose lead and corrosivity data were not available at the time of analysis.

TABLE XXX- EVALUATION OF MODELLED LARSON SKOLD AND HOUSING AGE INTERACTION DECISION
RULE IN REFERENCE TO LEAD DETECTION

Screening Classifications[a]	Below Detection (n=51)	Above Detection (n=46)	Row Total
Low Risk Homes[b]	43	29	72
High Risk Home	3	13	16
Total Used for Screening Tests	46	42	88
Missing Data for Screening Classification	5	4	9

Positive Predictive Value	*81.3%*
Negative Predictive Value	*59.7%*
Sensitivity	*31.0%*
Specificity	*93.5%*

[a] There was 1 sample with housing age data is missing corresponding lab data
[b] Risk criteria determined if the Larson Skold Index exceeded 0.59 and if the house of origin was built before 1986.

VI.   DISCUSSION

A. <u>Summary of Main Findings</u>

Nearly half of first draw water samples (n=47, 47.4%) had detectable levels of lead, though only 3 samples (3.1%) exceeding the EPA Action Level of 15µg/L.   As with other studies, we found first draw lead samples were significantly higher than last draw lead samples. The overall distribution of first draw lead was consistent across both counties, while last draw lead was significantly different by county. However, the latter was consistently quite low, with undetectable samples at even the 80th percentile.

Chloride, sulfate, and alkalinity were all significantly different between counties. Chloride levels were exceptionally elevated particularly in Kane County (Mean: 80.66 mg/L, Median: 36.8 mg/L). A few chloride samples in both Kane and Jackson counties (n=4, 4.1%) exceeded the EPA's secondary maximum contaminant levels (SMCL) of 250 mg/L, as well as the Illinois Pollution Control Board's promulgated standard of 500 mg/L (n=2, 2.1%) (Illinois Pollution Control Board, 2013; United States Environmental Protection Agency, 2015). Sulfate levels were elevated in Jackson County, with 1 sample (1.0%) exceeding the EPA SMCL of 250 mg/L.

While both corrosivity indexes used similar geochemical parameters, the Larson Skold had minimal variability between and within the two counties. The CSMR values observed, by contrast, varied much more by county, perhaps being driven more by local geology and land use (discussed in depth in the following section). In addition to Larson Skold's consistency, its established empirical thresholds were well distributed and had more utility as an ordinal predictor. There was a clearer stratification of lead detection and tertile status along these categories. By contrast, the decision rules for using CSMR to determine corrosivity, as developed in Nguyen et al (2010), did not seem particularly of use in our context. The bulk of samples, as classified by the CSMR, had "Significant Concern"-level corrosivity, which we interpreted as equivalent to a "moderate corrosivity" classification. Thus,

statistically significant associations (TABLE XIX) between the empirical thresholds and detection or upper

tertile status seemed to be a product of the commonality of the Significant category, indicating limited

utility of CSMR in documenting effects of corrosivity for our purposes. This should mean that rates of

galvanic corrosion (presuming that many of our wells had the applicable metal connections) should be

higher and, correspondingly, lead levels would be elevated. The rates of galvanic corrosion, therefore,

may require further investigation. We further evaluated CSMR as a predictor of WLL through sensitivity

analysis, finding a possible effect between Quartile 2 (0.24-1.4) and Quartile 1 (0-0.23), which is similar

to the previously defined CSMR threshold of 0.2. Further analysis did not reveal more compelling

evidence of increased lead detection.

We found that there was no significant relationship between housing age and corrosivity.

Specifically, it did not appear that housing quality and premise plumbing enhanced or was a predictor of

corrosive water behavior, and that geological setting and underlying housing stock conditions were not

coincident with each other. Premise plumbing and lead pipe can influence overall corrosivity through

becoming hosts for different microbes that may use different metals and dissolved species in processes

such as nitrification. Nitrification, in particular, can lower the pH of water and influence overall

conductivity (Zhang, Griffin, & Edwards, 2010; Zhang et al., 2009).

Our results indicated that there was a statistically significant (p=0.01) difference in the median

first draw lead levels by whether homes were constructed pre- and post-1986 (Pre-1986: 1.11 µg/L,

Post-1986: 0.54 µg/L (imputed ND value)). There were also greater proportions of detectable lead

samples in older homes than newer ones (Pre-1986: 58.6% Post-1986: 25.8%). Similarly, the odds of

lead detection were significantly greater in pre-1986 homes than in post-1986 homes (OR: 2.08, 95% CI:

1.68, 11.68). This is consistent with our expectations.

There were statistically significant associations between LSI and lead detection and with the presence of a lead level in the upper tertile. Lead was detected more frequently in samples with Moderate/High corrosivity. This was more pronounced when specifically looking at pre-1986 homes, where Low corrosivity samples were majority below detection and three times the proportion of the Moderate/High samples. Furthermore, we saw the same association using upper tertile lead with the Larson Skold thresholds (Above 67th: n=6, 26.08%, Below 67th: n=5, 14.7%). Our results suggest that there is indeed a significant positive association between elevated corrosivity and first draw lead in private well water, and that this association is accentuated by the age of housing for both detectable and above upper tertile lead. From our predictive modelling, we saw a significant stratum specific effect: the association between corrosivity (LSI>0.59) and water lead detection was stronger among pre-1986 homes (OR 4.13, 95% CI: 1.02, 16.68) than among homes built since 1986 (OR 2.22, 95% CI: 0.30, 16.56). However, statistical tests of interaction did not demonstrate an interaction between housing age category and corrosivity category on lead detection.   This is consistent with the large overlap in the confidence intervals of stratum-specific associations between corrosivity and lead detection for pre-1986 homes and post-1986 homes.   When we used the cutpoints of this stratum specific effect as a diagnostic screening rule in relation to lead detection on our data, we found that this tool had high specificity (93.5%) and positive predictive value (81.3%), but quite low sensitivity (31.0%) and low negative predictive value (59.7%).

What is less clear, however, is the magnitude and trend of this association. In all homes and in pre-1986 homes, corrosivity and lead did not exhibit a monotonic relationship according to Spearman's Rho Rank values. When corrosivity was evaluated as continuous in relation to detection status, median corrosivity values were typically higher for detectable lead samples than non-detectable lead samples, but these comparisons were not statistically significant. Similarly, when evaluating lead as a continuous variable with the Larson Skold empirical thresholds in pre-1986 homes, there was an indication of

increasing median lead concentrations from Low to Moderate/High, but this comparison was also not statistically significant. Results from our logistic regression modelling indicated the presence of cutpoints using the Larson Skold Index between the 3rd quintile (0.17-0.34) and the 1st quintile (0.01-0.05), as well as cutpoints between the 4th quartile (0.59+) and the 1st quartile (0.01-0.07). Only the association between the 4th and 1st quartile had a significant stratum specific effect in the presence pre-1986 housing. Our findings suggest that the use of 0.59 may be a more useful cutpoint for determining lead detection than some of the previously defined empirical thresholds, though as noted, this decision rule has low sensitivity in our setting, and would not be appropriate for public health screening purposes. However, additional investigation is required to rule out the possibility of a dose response relationship between lead and Larson Skold-measured corrosivity, particularly in relation to this threshold.

The three samples that exceeded the EPA Action Level (TABLE XXV) and our evaluation of the decision rule for screening (TABLE XXX) do complicate this picture, however. Two of the three were from homes constructed post-1986 (2011 and 2004), and all three had low Larson Skold corrosivity (0.01-0.23, within the Low category). All of them were largely composed of particulate lead, suggesting that this was more likely because of poor condition of a home's premise plumbing or a well's components. However, while older condition would make sense for specifically for the one pre-1986 home (JA-21: House Built: 1976, Well Construction: 1980), this would not for the other two samples (JA-36: House Built: 2011, Well Construction: unknown; KA-40: House Built: 2004, Well Construction: 2006). Furthermore, in the context of our data, our decision rule had a decent positive predictive value (81.3%), but a very low sensitivity (31.0%) relation to lead detection. Screening tests for lead need greater sensitivity, as the risks posed by lead exposure are serious enough to risk false positives and further analysis of water. Thus, more specific investigation in terms of premise plumbing and well condition is required to understand these very high WLL levels.

B. Comparisons with Related Studies

Study	Type	Location	Detection Limit	n	Differentiation of Bioavailable Forms of Lead	First Draw % Detect[e]	Mean (µg/L)	Median (µg/L)	90th %tile (µg/L)	Above Action Level	Second Draw/Flushed % Detect	Mean (µg/L)	M
Pieper et al 2018a.	CWS[b]	Flint, MI	1µg/L	268	Separate and combined quantification	85%	--	3.50	26.80	17.0%	47.0%	--	
Levallois 2014[a]	CWS	Montreal QC	0.01µg /L	306	Assessed in total lead quantification	--	--	--	--	--	--	--	
Edwards et al 2009	CWS	DC	--	47	No	--	--	--	90.00	--	--	--	
Pieper 2018b	PDWs[c]	Macon County, NC	1µg/L	15[d]	Separate and combined quantification	100%	247.8	30.00	--	67.0%	73.0%	5.70	
Pieper 2015	PDWs	VA	1µg/L	2144	Separate and combined quantification	--	22.00	4.00	--	19.0%	--	--	
Knobeloch	PDWs	WS	--	3868	No	--	--	--	--	--	--	--	
Swistock	PDWs	PA	--	251	No	--	--	--	--	12.0%	--	--	
Pieper 2018c	PDWs	Orleans NY	1µg/L	90	Assessed in total lead quantification	--	6.00	2 .00	17.00	12.0%	--	--	
Our Study	PDWs	IL	0.76 µg/L	97	Separate and combined quantification	47%	3.16	0.54[a]	5.00	3.1%	18.6%	1.86	

Our finding of statistically significant decreases between first and last draw lead are consistent with the literature, and particularly with the use of flushing as a mitigation method for reducing lead exposure. However, it is also worth noting that flushing it itself is not a reliable method, and water lead studies looking at private well supplied homes found observed sudden spikes in particulate lead after consistent flushing intervals (Pieper, Nystrom, et al. 2018; Katner et al. 2018; Levallois et al. 2014; Edwards, Triantafyllidou, and Best 2009). We also found that last draw lead levels were significantly different by county. Thus, even the use of first and last draw lead samples may lead to a mischaracterization of the issue, and may not be generalizable beyond the county level.

When looking at lead levels in comparison to other studies evaluating WLLs in private wells (*TABLE XXXI*), (Knobeloch et al., 2013; Pieper et al., 2015; Pieper, Nystrom, et al., 2018; Pieper, Tang, et al., 2018; Swistock et al., 2013) we find that a far smaller proportion of our first draw lead results exceed the action level than all the other studies. Median first draw lead was also drastically lower, as was

median second draw lead. The other private well studies also reported second draw lead action level exceedances, while ours did not have any. The study whose results most closely resemble our own are from a study conducted in Orleans, NY looking at the impacts of a nearby road salt storage facility combined with salt impacts from nearby major roads on water quality and private well corrosion (Pieper, Tang, et al., 2018). Even in that study, they recorded greater action level exceedances and a slightly higher distribution in first draw water lead.

Looking specifically at other community water system studies though (Edwards et al., 2009; Levallois et al., 2014; Pieper, Martin, et al., 2018), we found that the 90th percentile lead of our samples were lower than those found in Flint (Pieper, Martin, et al., 2018) and Washington, DC (Edwards et al., 2009). Our first draw median lead concentrations and proportion of lead detection were lower than Pieper, Martin, et al (2018). Our last draw median lead concentration was below detection in contrast to results from Montreal (Levallois et al., 2014), and our 90th percentile was much lower than values reported in Pieper, Martin et al (2018) and Levallois et al (2014). Thus, while we did find elevated levels and increased rates of detectable lead, they were much lower than other studies conducted for community water systems and private domestic wells.

The median chloride levels are higher than those found in Virginia, but are lower than those reported in Orleans, NY (Pieper, Tang, et al., 2018) and Flint (Masten, Davies, & Mcelmurry, 2016; Pieper, Tang, & Edwards, 2017) (**Error! Reference source not found.**). Our median sulfate concentrations are fairly similar to studies done in Flint, Virginia and New York, and exceed all of them. Both mean and median alkalinity levels are dramatically higher than in Flint. The Larson Skold average and median were both considerably lower than the measurements taken in Flint by the water utilities (Masten et al., 2016).   The median CSMR was lower than the median estimate calculated using utility monitoring data (Masten et al., 2016), but was similar to the average CSMR reported from more

extensive sampling (Pieper, Martin, et al., 2018). CSMR estimates are comparable with the results from

Orleans, NY, where 89% of wells in the study exceeded the 0.5 threshold (Pieper, Tang, et al., 2018).

Chloride levels, and thus the CSMR, are driven primarily by land use trends rather than

necessarily geology. Most groundwater in Illinois has relatively low levels of chloride and studies of

shallow groundwater in northern Illinois found that pristine aquifers ranged from <1 mg/L to 15 mg/L.

They concluded that level greater than 15 mg/L would be attributable to anthropogenic source (Kelly,

Panno, & Hackley, 2012; S. V. Panno et al., 2006). Thus, it is likely that land use trends in terms of

agriculture and heavily trafficked roads may drive trends in chloride. Road salt, fertilizers animal waste,

human waste from wastewater effluent, and landfill leachate all have had impacts on chloride levels of

ground water.

TABLE XXXI- COMPARISON OF PRIVATE DOMESTIC WELL AND COMMUNITY WATER SYSTEM STUDIES WITH WATER LEAD LEVELS

Study	Type	Location	Detection Limit	n	Differentiation of Bioavailable Forms of Lead	First Draw					Second Draw/Flushed (3 min flush)				
						% Detect[e]	Mean (µg/L)	Median (µg/L)	90th %tile (µg/L)	Above Action Level	% Detect	Mean (µg/L)	Median (µg/L)	90th %tile (µg/L)	Above Action Level
Pieper et al 2018a.	CWS[b]	Flint, MI	1µg/L	268	Separate and combined quantification	85%	--	3.50	26.80	17.0%	47.0%	--	--	6.60	4.0%
Levallois 2014[a]	CWS	Montreal QC	0.01µg /L	306	Assessed in total lead quantification	--	--	--	--	--	--	--	1.24	4.51	--
Edwards et al 2009	CWS	DC	--	47	No	--	--	--	90.00	--	--	--	--	--	--
Pieper 2018b	PDWs[c]	Macon County, NC	1µg/L	15[d]	Separate and combined quantification	100%	247.8	30.00	--	67.0%	73.0%	5.70	2.00	--	7.0%
Pieper 2015	PDWs	VA	1µg/L	2144	Separate and combined quantification	--	22.00	4.00	--	19.0%	--	--	--	--	--
Knobeloch	PDWs	WS	--	3868	No	--	--	--	--	--	--	--	9	--	1.8%
Swistock	PDWs	PA	--	251	No	--	--	--	--	12.0%	--	--	--	--	--
Pieper 2018c	PDWs	Orleans NY	1µg/L	90	Assessed in total lead quantification	--	6.00	2 .00	17.00	12.0%	--	--	--	--	--
Our Study	PDWs	IL	0.76 µg/L	97	Separate and combined quantification	47%	3.16	0.54[a]	5.00	3.1%	18.6%	1.86	0.54*	1.22	0%

[a] First sample taken was let to flush for 5 minutes, so most comparable to last draw

[b] CWS=Community Water System

[c] PDWs=Private Domestic Wells

[d] Small pilot sampling

[e] 0.54 is the imputed value of the limit of detection by calculating $\frac{Sample\ Quantification Limit}{\sqrt{2}}$

Specifically, in the context of the Chicago area, trends in road salt use and proximity to major roads have been associated with increased chloride levels of shallow aquifers (Kelly, 2008), while agricultural inputs are more important in more rural areas (Kelly et al., 2012).

TABLE XXXII-COMPARISON OF GEOCHEMICAL AND CORROSIVITY PARAMETERS TO OTHER STUDIES

Location	Study	Type of Study	Chloride (mg/L)		Sulfate (mg/L)		Alkalinity (mg/L)		Larson Skold		CSMR	
			Mean (Range)	Med. (Range)	Mean (Range)	Med. (Range)	Mean (Range)	Med. (Range)	Mean (Range)	Med. (Range)	Mean (Range)	Med. (Range)
Flint, post water switch	Masten et al 2016[a]	CWS[1]	79.7 (62-95)	83.0 (62-95)	24.5 (21-31)	24.0 (21-31)	65.5 (36-118)	58 (36-118)	2.50 (1.24-3.40)	2.58 (1.24-3.40)	3.32 (2.8-3.8)	3.35 (2.8-3.8)
	Pieper et al 2018a	CWS	80.0 (74-87)	--	39.2 (24.3-45.0)	--	--		--		2.04	--
Orleans NY	Pieper et al 2018c	PDW[2]	179.3	89.5	30.7	27.0	--		--		Above 0.5: 89%	
Virginia	Pieper et al 2015	PDW	14.3	5.4	45.1	8.0	--		--		--	
Illinois	Our study	PDW	68.5 (0.8-1022)	25.1 (0.8-1022)	40.8 (0.2-319.0)	29.3 (0.2-319.0)	338 (65-725)	344 (65-725)	0.41 (0.01-2.24)	0.23 (0.01-2.24)	40.5 (0.04-1091.0)	2.1 (0.04-1091.0)

[a] Results from Masten et al (2016) are derived from 5 presented data points in their appendices

[1] CWS=Community Water System

[2] PDWs=Private Domestic Wells

Similar to other studies, we found that older, pre-1986 homes had significantly higher water lead levels than newer homes (

C. **Strengths of Study**

A key strength in this study is the more precise and careful consideration given to evaluating and quantifying corrosivity. Previous studies evaluating water lead levels assumed that corrosivity was merely a product of pH, with a few using CSMR. By contrast, our use of both CSMR and Larson Skold

gave our study more precise explanatory power in understanding the mechanisms involved in lead exposure at the tap, as well as how these metrics behave in the context of private well testing. From our results, we found a more consistent association between Larson Skold-related cutpoints and elevated lead rather than with CSMR. This also enabled us to analyze the influence of chloride levels, which is a remarkable finding in itself. The interplay of chloride levels in relation to private well water corrosion and water lead has only been analyzed recently (Pieper, Tang, et al., 2018).

Another strength of our study is being very clear about how to conduct the sampling. We were very intentional about how sampling was conducted, and explicitly had participants take first and last draw samples in order to evaluate both the impacts of premise plumbing and the well components on water lead. This allowed us to properly analyze effects resulting from both premise plumbing and well components, as demonstrated through our stratified analyses with pre-1986 homes and our logistic regression modelling results.

TABLE XXXIII). This phenomena has been observed in other studies conducted on private domestic wells in PA and VA (Pieper et al., 2015; Swistock et al., 2013), and community water systems in New Orleans (Katner et al., 2018). Our samples came from older homes than Pieper et al (2015), although the range of housing ages for both studies was quite similar. The percentage of older homes with detectable lead was lower but comparable to percentages reported in Pieper et al (2015) and Katner et al (2018). The magnitude of difference between pre and post 1986 homes (Medians: Pre: 1.11 µg/L, Post: 0.54 µg/L (imputed ND); p=0.014) was lower than differences re reported in Pieper et al (2015) (Medians: Pre 1988: 5.4 µg/L, Post 1988: 3.3 µg/L). Katner et al (2018) reported results of multiple variable logistic regression modelling. They found that, after adjusting for occupancy and flush time, the odds of pre 1950 homes having detectable lead levels were 2.95 times higher than post-1950 homes (95% CI: 1.80, 4.83). Our own results indicated, in univariate models, that the odds of detectable lead in pre-1986 homes vs post-1986 homes were 2.08 times higher (95% CI: 1.68, 11.68), but our

modelled odds with and without stratum specific effects with different transformations of Larson Skold generally exceeded those of Katner et al, (2018).

D. **Strengths of Study**

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TABLE XXXIII- COMPARISON OF LEAD LEVEL BY HOUSING TO OTHER STUDIES

							Pre-Post Comparison	
Study	Location	Type of Study	n	Detection Limit (µg/L)	Average Year/Distribution	% Before With Detectable Lead	Median Differences (µg/L)	Odds Ratio

Swistock et al 2013	PA	PDWs	251	--	--	Elevated WLL Homes built before 1991: 70% had copper plumbing installed	--	--
Pieper 2015	VA	PDWs	1405	1.00	1988 (1850-2013)	Before 1988: 42.70%	Pre 1988: 5.4 Post 1988: 3.3 Change: 2.1*	--
Katner et al 2018	New Orleans, LA	CWS	325	1.00	66% pre 1950 17.6% post 1950	Pre 1950: 73.4% (after flushing)	--	Adjusted for flush time and occupancy Pre-1950: 2.95* (1.80, 4.83)
Our study	IL	PDWs	89[c]	0.76	1974 (1850-2013	Pre 1950: 6.74% Pre 1986: 38.20% Pre 1988: 39.33% Pre 1991: 40.45%	Pre-1986 : 1.11 Post-1986: 0.54[b] Change: 0.57*	LSI (Quartile 4), pre-1986: 4.13* (1.02, 16.68)

[a] Referent is post-1950 buildings and homes
[b] Imputed value for ND
[c] There are 9 samples that do not yet have survey data available
*Significant at α=0.05

Our study appropriately captures multiple forms of ingestible lead, and evaluated the ingestible lead speciation (particulate vs. soluble) from a limited amount of samples. We only analyzed samples that exceeded the 15 µg/L action level for composition of dissolved lead. This was somewhat useful in understanding our three major exceedances, but evaluating source composition of remaining samples would help better understand how corrosivity is driving detectable and elevated water lead levels.

Finally, our study more explicitly collaborated with LHDs in terms of recruitment, training and implementation. We believe that this arrangement helps make this research more translational by providing a model for others looking to evaluate private well water lead. It also sets up potential follow-up through having a partner to discuss possible interventions.

E.   Limitations of Study

One of the limitations of our study comes from how we picked our counties of study. We ended up focusing on the interest of local health department in participating in the study, rather than picking counties based on specific characteristics, such as demographics or geological considerations. Nevertheless, the research was conducted in one county in the northern, one in the central, and one in the southern part of Illinois. While initially we hoped to have a geographically representative sample of each county (and LHD staff strived for this), we ultimately were focused on understanding the worst case scenario by focusing on older homes. Nonetheless, this still did limit the generalizability of our study.

Another limitation was our cut-off for determining initial eligibility, as it might have been too conservative. We used 1978 as the cutoff year for eligibility, which corresponded to the year that the CPSC lead paint ban went into effect. This was a product of earlier intentions of the study, which incorporated lead paint inspections as part of in home sampling. However, in our analysis, we used pre-1986 as our cut off for determining old vs new houses. This may have skewed differences in lead levels, as some of the newer houses within the range of 1978-1986 may have had lower lead levels. Nonetheless, this study produced similar distributions of housing stock age across both counties, with a large fraction of the distribution within the aforementioned window. This is encouraging for future research that might expand the scope of this study.

Our reliance on participant expertise, recall, and implementation may have also affected study outcomes as well. LHD staff trained participants in their homes about water sampling, but we were reliant on participants to appropriately take samples and label them. This could have resulted in negative confounding between differences in first and last draw lead. Mixing the first and last draw samples would have also affected the geochemical parameter analyses, as these analyses were carried out specifically on last draw samples, presuming that flushing before sampling would yield

measurement pertaining to well conditions. Data on the age of the house may have also been affected

by our reliance on participants. About 9 participants (9.2%) could not recall the age of their house, and

others may have misremembered these data.

Sample size was a limitation for our analysis, and we had to collapse categories to improve

power in analyses of corrosivity categories. While we did find a significant association even with our

small sample size, there may have been additional effects which we simply did not have the power to

detect. Specifically, a greater sample size may have helped with some of the single predictor logistic

regression models in terms of Larson Skold, and in better teasing apart the effects of CSMR on lead

detection.

As alluded to in the summary of corrosivity findings, this study did not observe or analyze land

use trends and their impacts on both corrosivity and water lead levels. Both counties have a significant

amount of agricultural land whose fertilizer and chemical run off may elevated chloride levels, and we

did not geolocate our wells near them. As of 2017, both counties have multiple acres of farmland, with

Jackson County (221,621 acres) having 30% more farmland than Kane County (170,254 acres) and 40%

more farms (Jackson: 687 farms, Kane: 490 farms) (United States Department of Agriculture, 2017).

Road salt run-off, particularly from major, well trafficked roads, can impact chloride groundwater levels.

These impacts have been observed particularly in the eastern portion of Kane County (Kelly, Hadley, &

Mannix, 2016). The impacts of increased road salt-originating chloride on groundwater corrosivity was

specifically investigated in Orleans, NY (Pieper, Tang, et al., 2018). In this study, the researchers

evaluated whether a leaking salt barn (classified as "Salt Barn") or being within 30 m to a major ("Major

Road") or neither ("Minor Road") was influencing the water quality of private drinking water wells.

Spatial trends indicated elevated CSMR levels clustered around a leaking road salt barn of interest and

by a major intersection. The researchers found a significant negative correlation ($\rho = -0.40$) between a

well's distance from the salt barn and measured chloride. Statistically significant differences,

particularly between the Salt Barn and the Minor Road wells, were observed for chloride levels and CSMR values, with increasing median chloride level for each well depending on each grouping. It is interesting to note, however, that lead concentrations did not vary significantly based on a well's distance from the salt barn or on the above classifications. The researchers attributed this to differences with premise plumbing composition. Additionally, median lead levels were significantly higher when CSMR values were greater than 0.5 ("Significant concern") (Wilcoxon Test, $p < 0.05$), and additional bench testing conducted as part of this study found that samples where chloride levels exceeding 250 mg/L had significant median increases in lead leaching from lead solder joints. Thus, particular land use factors can impact both the chloride and CSMR measurements of nearby wells, and could potentially influence lead levels themselves.

Jackson County's history of coal mining may have also had an influence on the sulfate levels of nearby wells. A majority of these mines (surface and underground) have been abandoned, although there are still a few active mines. Areas with both active and abandoned surface and underground coal mines have altered stratigraphy and hydrogeochemistry that may impact groundwater corrosivity and acidity. This has been observed in Illinois with both underground mining (Booth & Bertsch, 1999) and, to a lesser extent, with surface mining (Lindorff, Cartwright, & Herzog, 1981).

VII.   CONCLUSIONS

A. <u>Public Health Implications</u>

Local health departments in rural areas, particularly areas where private domestic wells are common, may be interested in testing and surveillance of water lead levels. These often have financial and personnel constraints, and thus may need a way to triage high priority homes and sub-populations. Our results suggest that that there is a significant stratum-specific effect between age of housing and increased corrosivity, using 0.59 as a threshold for Larson Skold measurements. Additionally, much of this corrosivity is driven by the presence of chloride levels. Thus, LHDs looking for cost effective ways of conducting surveillance could focus on pre-1986 homes and try to identify areas more likely to have corrosive water according to this cutpoint. However, the specific decision rules we employed – pre-1986 housing age and Larson Skold index corrosivity ≥0.59 – had low sensitivity. Thus, for homes with domestic wells and individuals in life stages that make them more susceptible to serious lead impacts (i.e. pregnant mothers, infants, and children) – testing water regardless of housing age and corrosivity may be appropriate. Developing more expertise in evaluating premise plumbing and well components may further help in this regard to more precisely determine if lead levels might be elevated in specific homes.

Our results for the Illinois pilot represent a more attenuated trend in relation with the other studies conducted in Virginia, Wisconsin, Pennsylvania, North Carolina, and New York. This consistency in trend, if not in magnitude, makes evident the case for evaluating lead in private drinking water wells in Illinois. While our pilot study largely found that water lead levels are not as elevated as in other locations, no level of lead is safe. Even if one were to assume that corrosive groundwater-driven lead contamination is less of a concern in Illinois, this would ignore the unpredictability of particulate lead fluxes. Just relying on the Larson Skold cutpoint we established would ignore the three samples that exceeded the EPA Action Level. These three samples did not have high corrosivity, and two of the three

were not from older homes. Nonetheless, this focus is appropriate and feasible given the limitations of LHDs. To overcome the possible limitations of this corrosivity-oriented approach, developing expertise in evaluating well and plumbing system composition can enhance a LHD's ability to address this issue.

From a policy standpoint, the presence of a possibly large scale water quality issue may necessitate the need for LHDs and the state of Illinois to develop or invest in cost effective regulation interventions. This is important because, according to Illinois BRFSS, the majority of well owners make under $50,000 a year, may not have a high earning potential (~40% have at most a high school diploma), and own their own property. This is on top of the financial limitations of LHDs in rural areas. In terms of testing, using a similar suite of parameters as shown in this study could appropriately evaluate water lead levels. However, though cheaper than other analytical testing programs, the $40 per first and last draw sample may still be burdensome without state subsidy. It is also a challenge of elective water quality testing to get people to understand their results and subsequently act on the resulting data. Programs in Pennsylvania (Pennsylvania Master Well Owner Network) and Virginia (Virginia Household Water Quality Program and Virginia Master Well Owner Network are examples of state extension programs that combined subsidized testing, testing education and interpretation, with recommendations with how to proceed in terms of maintenance and installation of treatment devices (Benham, Ling, Ziegler, & Krometis, 2016; Clemens, Swistock, & Sharpe, 2007). These programs may still not reach socioeconomically disadvantaged individuals who are most at risk, as seen in studies conducted in New Jersey. Even when free testing programs are offered with community outreach programs, these are often still utilized by individuals with higher socioeconomic status, exacerbating current inequities (Flanagan et al., 2016), Based on this research involving arsenic, Zheng and Flanagan (2017) argue that universal testing policies would be more effective than anything at the state level.

B. Future Research Directions

While we have evaluated the phenomenon writ large, necessary follow-up would need to

include defining areas that are more prone to corrosive water and possible contamination. This may be

done as part of identifying at risk areas based on proximity to potential sources, as well as the risk

factors that might contribute to increased water corrosivity. Road salt particularly has been implicated

in shallow aquifer contamination in the northern part of the state (Kelly, 2008). Thus, future research

may evaluate contamination source mitigation and prevention as well.

The nature of the Larson Skold Index relationship to elevated water lead is still an open question

that needs further investigation. While we analyzed and found a cutpoint different from pre-established

empirical thresholds, it still would require greater validation to be more fully deployed. Additionally,

further research is required to evaluate the strength and slope of association between corrosivity

metrics and elevated water lead to further define if corrosivity operates by a dose response relationship.

This would also extend to CSMR in the context of private drinking water well corrosion. This metric has

really only been used in few studies with regard to groundwater and private domestic wells (Pieper,

Tang, et al., 2018; Tang et al., 2018), but did not seem to have much utility in our study. Nonetheless,

the elevated chloride levels, and the different types of corrosion measured by CSMR may require further

analysis for its utility.

Thirdly, our research used housing age as a proxy for premise plumbing and well condition, but

this may not necessarily be a concise proxy. Currently, a Phase II of this project has been planned,

where homes with elevated water lead above the EPA Action Level will receive testing for a broader

array of contaminants. However, better understanding materials-related insights could help to triage

which homes need to be more closely evaluated. Additionally, evaluations of well components would

shed greater light on trends in the corrosive water-to-elevated water lead mechanism, which would aid

in addressing this issue from a public health standpoint. Such information may result in improvements in

the sensitivity of predicting the presence of measureable lead in tap water of homes with domestic wells.

Fourthly, future studies need to investigate whether at-risk populations, such as pregnant and breastfeeding mothers, infants, and children, are consuming contaminated well water, and how much. We are still unsure about the extent to which individuals are actively consuming their well water without any sort of exposure mitigation technology or strategy. If well water is simply not being drunk or is filtered before vulnerable populations consume them, then this is far less of an issue. Some dimensions to address of this include BLL testing, as well as the internal dose individuals may be consuming.

Finally, an evaluation of cost-effective interventions, as well as the financial and logistical feasibility of mandatory testing polices, are essential to following up on this research. Since well-owners are personally responsible for well maintenance and LHDs are constrained by their budgets, any successful intervention or outreach needs to be specific and quite targeted. This may also include directing homeowners to federal or state programs that may be available for well improvements. At the federal level include the USDA's Single Family Housing Repair Loans & Grants program (United States Department of Agriculture, 2019), and various state level programs such as the ones offered in Minnesota and Wisconsin (Minnesota Department of Health, 2019; Wisconsin Department of Natural Resources, 2019).

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