Investigating Lead Corrosion in Drinking Water Systems

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Abstract

In 1986, Congress banned lead components in drinking water distribution systems due to the harmful health effects of lead; however, existing lead pipes are still in use. Pipe loop and coupon rack tests to measure the corrosivity of water are time-intensive and expensive. The team evaluated a short-term laboratory test for predicting corrosion and found evidence of corrosion within one week for samples with extreme pH conditions (3 or 12). Samples with moderate pH and alkalinity conditions expected in a distribution system showed minimal corrosion after four weeks, and dissolved lead concentrations were inconsistent. Therefore, current testing techniques are recommended to differentiate corrosion potential in typical water systems and protect public health.

Executive Summary

Lead poses serious health risks, especially in newborns, children under 6, and pregnant women. Any amount of lead in the human body is unsafe and can cause damage to the nervous system, which can lead to learning disabilities. When lead exceeds 10 µg/dL in the blood, it can be fatal in children. In concentrations higher than 3 µg/dL, it can cause infertility. At extremely high levels, those higher than 30 µg/dL, it can cause miscarriage. Before the health risks were known, lead was used in pipes and distribution system components to deliver water to households, because lead is both malleable and durable. The use of lead pipes has led to scenarios where lead leaches into the water and is then consumed resulting in various health effects. The use of new lead pipes and components was banned in 1986, but existing lead components were allowed to remain in systems. Legislation aimed at controlling concentrations of dissolved lead in potable water is based on monitoring of lead concentrations and implementation of optimized corrosion control strategies.

The pipe-loop test and coupon test are used to assess the likelihood that water will corrode lead pipes and components. The pipe loop test is conducted in a laboratory setting with a section or multiple sections of pipe isolated. The pipes can either be new or cut from an existing system. The pipes are made into a loop, and water representative of the distribution system water is then circulated through each section constantly. While variables can be controlled to see the impact of changes, this test usually takes a minimum of six months, but is often run for one or more years. Because of the length of testing time, these tests are costly to conduct. A coupon test uses lead coupons mounted inside a rack, which is a length of PVC or similar pipe mounted in a back-and-forth pattern on a solid backboard and connected to a water system. The metal strips are typically exposed to water flowing through the coupon rack for 60, 90, or 120 days. After removal from the coupon rack, the metal strips have their initial weight compared to the final weight, and the difference in weight is used to estimate the rate of corrosion. This testing method only reflects the conditions at the site of the coupon rack, resulting in temperature having little effect on results even though temperature plays a large part in corrosion.

The goal of this project was to determine if a short-term laboratory test is a viable option to predict lead corrosion. A short-term test could replace current testing methods or be useful in narrowing the scope of investigation before current methods are used. Specifically, this project evaluated corrosion of coupons in a bench-scale test performed over four weeks using water samples with varying pH, alkalinity, and temperature.

The laboratory test involved the creation of water samples with varying alkalinity (10 to 200 mg/L as $CaCO_3$) and pH (3.02 to 8.29). Lead coupons were placed in each sample and the samples incubated at 4 or 20°C. Aliquots were removed for lead concentration testing at different times over a four-week period. At the conclusion of the testing period, dissolved lead, pH, alkalinity, and lead coupon weight were measured for each sample. Lead was measured using an inductively coupled plasma-optical emission spectrometer (ICP--OES), pH was measured using an Accumet basic AB15 pH meter, alkalinity was measured by volumetric titration with HCl, and weights were taken using a Mettler Toledo MS104TS/00 analytical balance.

Of the 80 samples tested, 17 samples had detectable concentrations of lead while 63 aliquots were below the detection limit (BDL). For the detectable concentrations, 12 occurred in strong acid or strong base solutions, and the other five occurred in samples where the alkalinity was the independent variable. For the acidic and basic solutions, lead was detected as early as the day one test, indicating that the solutions quickly were leaching lead, and levels rose over the course of the test. For example, the sample with pH of 3 at a temperature of 20°C had 5.55, 95.0, 118, and 111 µg/L of lead after 1 day, 1 week, 3 weeks, and 4 weeks, respectively. These low pH conditions and high lead levels in the hundreds of µg/L would not be expected in a water distribution system, for which the action level for lead is 15 µg/L as a 90th percentile value. For samples with conditions more typical of a distribution system, such as the sample at 20°C and alkalinity at 100 mg/L as $CaCO_3$, the dissolved lead concentrations were below detection limits (BDL) for the 1 day, 1 week, and 3-week samples, and 1.31 µg/L after 4 weeks. The samples with alkalinity did not have detectable levels of dissolved lead until the third and fourth weeks, indicating that those samples took longer to corrode.

Results demonstrated that a short-term, bench-scale corrosion potential test can detect corrosion in waters with extreme conditions, such as those with extremely low or high pH values. However, such conditions would not be expected in a water distribution system. For conditions that are expected to minimize corrosion, such as waters with near neutral to slightly alkaline pH values and moderate alkalinity concentrations, a short-term test was not able to differentiate between samples as most had non-detectable concentrations of dissolved lead. Extending the test time frame may yield more results in such solutions. In addition, measuring lead concentrations on an instrument with a lower detection limit, such as an inductively coupled plasma mass spectrometer, may provide more useful data. At this time, the pipe loop or coupon rack test may be a more reliable test for differentiating lead corrosion potential in typical water distribution systems. While these tests are time-intensive and expensive, the investment is warranted to protect public health.

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1. Introduction

In 1986, lead pipes and other lead components were banned in drinking water distribution systems by Congress. However, pipes installed before 1986 may still be in use (Cornwell et al., 2016). In 2016, two surveys sponsored by the American Water Works Association (AWWA) found that 30% of community water systems reported having lead service lines in their system. These results were used to calculate a national estimate of 6.1 million lead service lines nationwide, with a lower bound of 5.5 million and an upper bound of 7.1 million (Cornwell et al., 2016).

Lead in drinking water can cause negative human health effects, especially in children and pregnant women. Signs and symptoms of lead poisoning include developmental delays, learning difficulties, irritability, loss of appetite, weight loss, sluggishness, fatigue, abdominal pain, vomiting, constipation, hearing loss, seizures, and pica, an eating disorder in which the affected person eats things that are not normally considered food. At extremely elevated levels (10 µg/dL blood level), lead poisoning can be fatal (Needleman & Landrigan, 2004). Another common component of water conveyance systems is copper, which is an essential nutrient but can have negative health effects at high concentrations.

The Safe Water Drinking Act (SWDA) was established in 1974 and then revised in 1986 and 1996, to protect the public and maintain safe standards for public drinking waters (U.S. Environmental Protection Agency, 2004). The SDWA requires the Environmental Protection Agency (EPA) to set limits for contaminants that can have adverse health effects. First, the EPA determines levels at which contaminants in drinking water would have no adverse health effects, known as maximum contaminant level goals (MCLGs). Next, the EPA sets an enforceable standard that is as close to the MCLG as possible, taking into consideration technical and economic feasibility. Depending on the contaminant, the enforceable standard is a maximum contaminant level (MCL), a treatment technique (TT), or action level (AL). A maximum contaminant level (MCL) is the highest level a contaminant can be in drinking water and is often set as close to the MCLG as possible (U.S. Environmental Protection Agency, 2022e). An action level is a level at which provisions laid out in the rule must be initiated by the water system (Centers for Disease Control and Prevention, 2017). The EPA has set the MCLG for lead at zero, due to lead’s toxicity and bioaccumulation characteristics, and the MCLG for copper at 1.3 mg/L due to the possibility for liver and kidney damage at high exposure (U.S. Environmental Protection Agency, 2022c). In addition to the MCLGs, the Lead and Copper Rule created an action level of 0.015 mg/L for lead and 1.3 mg/L for copper (U.S. Environmental Protection Agency, 2008).

Currently, there are two common tests for determining the corrosivity of a water in a pipe: pipe loop testing and coupon testing. For a pipe loop test, pipes are made into a loop and water representative of the distribution system water is then run through each section constantly. This test usually takes a minimum of six months, but is often run for years, making it costly to conduct (Civardi & Gray, 2005). Coupon testing involves strips of metals that are present in a particular piping system. The coupons are placed in a coupon rack made of a non-corrosive material (such as PVC). The coupons are typically exposed to water flowing through the coupon rack for 60, 90, or 120 days. They are then removed, and the weights are analyzed to determine the rate of corrosion. This testing method only reflects the conditions at the site of the coupon rack, resulting in temperature having little effect on results even though it plays a large part in corrosion (Welsh, 2021).

The purpose of this project was to examine if a short-term laboratory test could produce results to demonstrate if certain water conditions would be corrosive. Water solutions were created with different pH and alkalinity values; lead coupons were placed inside each sample; and the samples were incubated at different temperatures. Typical pH and alkalinity values were tested, as well as extremes. Aliquots were removed after 1 day, 1 week, 3 weeks, and 4 weeks and analyzed for dissolved lead concentrations. After a month of incubation, the coupons were removed from the water solution and weighed for analysis.

2. Background

2.1 Corrosion Chemistry

Corrosion is a chemical process where metal reacts with water, resulting in the dissolution of the metallic atom (The Electrochemical Society, 2022). In this process, the metal substance is oxidized in an oxidation-reduction (redox) reaction, in which electrons are lost by the substance that is oxidized and gained by the substance that is reduced. In both cases, the oxidation numbers of the oxidized and reduced molecules are changed (Nivaldo, 2018). In order for corrosion to occur, all of the components of an electrochemical cell must be present. This includes an anode, a cathode, a connection between the two, and an electrolyte solution. An anode is “the electrode in an electrochemical cell where oxidation occurs,” while a cathode is “the electrode in an electrochemical cell where reduction occurs” (Nivaldo, 2018). The electrons move from the anode to the cathode. Therefore, the cathode is an electron acceptor, and oxygen is the most common electron acceptor because of its high oxidation potential. If oxygen is not present, another high-energy electron acceptor would be required such as nitrate. An example of a redox reaction is shown in Reaction 2-1:

$$2Fe(s) + O_2(g) + 2H_2O(l) \rightarrow 2Fe(OH)_2(s) \qquad \text{(Reaction 2-1)}$$

This example redox reaction is the process of rusting, in which the iron is corroded by the water. In this example, the anodic half-reaction involves oxidation of the iron and is shown in Reaction 2-2, and the cathodic half-reaction in which oxygen is the electron acceptor is shown in Reaction 2-3:

$$2Fe(s) \rightarrow 2Fe^{2+}(aq) + 4e^- \qquad \text{(Reaction 2-2)}$$

$$O_2(g) + 2H_2O(l) + 4\ e^- \rightarrow 4OH^-(aq) \qquad \text{(Reaction 2-3)}$$

In drinking water distribution systems, copper and lead pipes, fittings, and the metals in solder can corrode. Example reactions for copper corrosion in which chlorine is the electron acceptor are shown in Reactions 2-4 through 2-6, including the overall, anodic, and cathodic reactions, respectively.

$$2Cu(s) + Cl_2(aq) \rightarrow 2CuCl(s) \qquad \text{(Reaction 2-4)}$$

$$2Cl^-(aq) \rightarrow Cl_2(g) + 2\ e^- \qquad \text{(Reaction 2-5)}$$

$$Cu^{2+}(aq) + 2\ e^- \rightarrow Cu \qquad \text{(Reaction 2-6)}$$

Similarly, in a household with lead pipes, fittings, or solder that has corroded, lead would be the anode and oxygen would be the cathode.

The tendency of a metal to corrode is determined based on the galvanic series, as seen in Table 1. The galvanic series is “a list of metals and alloys based on their relative potentials,” where metals with the most negative potentials are the most active, and metals with the most positive potentials are the least active (most noble) (Ahmad, 2006). The series is used to determine what metal will corrode when two or more are present in an electrically charged environment. For example, if both lithium and copper were present in an electrically charged environment, according to the galvanic series, lithium is more likely to corrode because it has a more negative potential. In addition, the corrosion potential is higher when the difference in potential between two metals is higher.

Table 1. The Galvanic Series (Shackelford et al., 2015)

Metal	Potential, volts (V)
Anodic or corroded end	
Lithium	-3.04
Rubidium	-2.93
Potassium	-2.92
Barium	-2.90
Strontium	-2.89
Calcium	-2.80
Sodium	-2.71
Magnesium	-2.37
Beryllium	-1.70
Aluminum	-1.70
Manganese	-1.04
Zinc	-0.76
Chromium	-0.60
Cadmium	-0.40
Titanium	-0.33
Cobalt	-0.28
Nickel	-0.23
Tin	-0.14
Lead	-0.13
Hydrogen	0.00
Copper	0.52
Silver	0.80
Mercury	0.85
Palladium	1.00
Platinum	1.20
Gold	1.50
Cathodic or noble metal end	

While corrosion can occur between any metallic substance and water, two metals of particular interest in drinking water distribution systems are lead and copper. These two metals are used in the conveyance of water to consumers, both in drinking water distribution systems and inside homes, and are regulated due to potential negative health effects. Lead was historically used in service lines, household plumbing, fixtures, and solder dating all the way back to ancient civilizations because it was readily available, malleable to work into different shapes, and unlikely to spring a leak (Plumbing Manufacturers International, n.d.). However, its use is banned or limited in modern construction due to potential negative health effects which are discussed in Section 2.1.3. Copper is used in household plumbing. The following sections provide details on materials used in water piping systems, and factors affecting corrosion of those materials.

2.1.1 Metals Used in Water Distribution Systems

Water distribution systems consist of many components including pipelines, storage facilities, and pumps. Pipelines, which are also called water mains, convey water from the untreated source or water treatment plant to different regions of a service area, and these pipelines are categorized based on size and purpose. Transmission mains are larger diameter pipes (typically larger than 3 feet) that transport water from the water source to feeder mains or storage facilities. Storage facilities include standpipes, reservoirs, and ground-level or elevated tanks that are used to provide water during periods of high demand and for firefighting. Feeder mains are smaller diameter pipes (less than 3 feet) and transport water from the water source to storage tanks. Distribution mains are small to mid-sized pipes (less than 2 feet diameter) and distribute water from a feeder main to a local service area (Wilke, 2018).

Primary feeders transport a high volume of treated water from the water treatment plant to a specific service area in the system. If the water distribution system is large, there may be multiple primary feeders to service different areas. The water from the primary feeders is then split into smaller pipes or mains to be distributed locally (National Research Council (US) Safe Drinking Water Committee, 1982).

Water mains are typically made of cast iron, steel, reinforced concrete, ductile iron, plastic, or asbestos-cement. In 2002, a distribution survey was sent to 3,000 water utilities asking questions about their distribution system. While the response rate was only 11%, these data were used to estimate the national pipe inventory. The survey found the pipe compositions were: 19.7% cement mortar lined ductile iron, 16.6% polyvinyl chloride, 15.2% asbestos cement, 14.4% cast iron, 14.4% cast iron unlined, 4.3% unlined ductile iron, 3.8% steel, and the remaining unknown/other (U.S. Environmental Protection Agency, 2007).

The next component of a water distribution system is the service connection, which is depicted in Figure 1. The service connection joins the water main to individual homes or buildings. Next to the water main is a gooseneck, which is an S-shaped piece of pipe that is flexible, to allow movement of the service or water main without the connection being lost. Goosenecks may be made of lead due to lead’s flexibility which allows for ground movement during different seasons, such as frost heaves in winter. The water flows through the gooseneck to the curb stop (City of Viroqua, 2022). A curb stop is a water service shutoff valve that is typically in the piping near the curb, between the building and water main. This valve is used to start or stop the flow of water from the main water line. The water then enters a water supply pipe that brings the water to the water meter, which measures the amount of water being used by the building so that the customer can be billed accordingly. The water then goes to an isolation valve. Isolation valves control the flow of water and are typically used for maintenance or safety purposes to stop the flow of water (Jetset Plumbing, n.d.). The water then enters the building and travels through the internal plumbing.

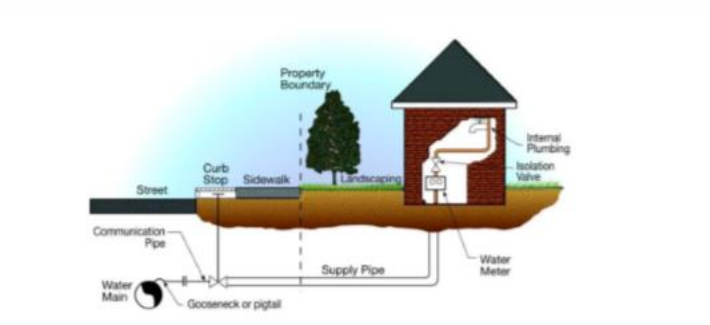

Figure 1. A Typical Water Service Connection (U.S. Environmental Protection Agency, 2016)

Service lines are typically made of galvanized iron, lead, galvanized steel, plastic, copper, ductile iron, or cast iron, with copper being the most widely used (National Research Council (US) Safe Drinking Water Committee, 1982). The service line could be owned by the water system, the property owner, or both (Neltner, 2021). Lead pipes were banned by Congress in 1986; however, pipes that were installed before then may still be in use. In 2016, two American Water Works Association (AWWA) sponsored surveys of U.S. community water systems (CWSs) were conducted to determine the occurrence of lead service lines (LSLs) in regions of the United States. The results found that 30% of CWSs reported having some LSLs in their system. The results of the two surveys were combined to calculate a national estimate of 6.1 million LSLs nationwide, with a lower bound of 5.5 million and an upper bound of 7.1 million (Cornwell et al., 2016).

Researchers have found that lead service lines contribute a significant amount of lead in standing samples collected at a tap, and that brass may also contribute lead and copper, depending on the quality of the water (U.S. Environmental Protection Agency, 2016). To combat this, there are federal and non-federal funding sources to help states and water utilities to replace lead service lines. Funding sources include the Drinking Water State Revolving Fund, HUD Community Development Block Grant, Assistance for Small and Disadvantaged Communities Grant, and the Reducing Lead in Drinking Water Grant. While LSL replacement programs permanently remove lead pipes from service connections, lead corrosion products that deposited onto other non-lead pipes in the system can still leach lead even after the LSL is replaced. However, LSL replacement programs are an important component of lead control practices in a system (U.S. Environmental Protection Agency, 2022f).

2.1.2 Metals Used in Premise Plumbing

Premise plumbing is the portion of the distribution system from the main water meter to the consumer’s tap in a home, school, or other building (U.S. Environmental Protection Agency, 2022b). In a home, the main supply is typically piped to the water heater, where it is divided into hot-water and cold-water pipes. From the water heater, supply pipes travel away in pairs – one hot and one cold – to distribute water throughout the house (Vandervort, 2021).

The five most common materials used in premise plumbing are polyethylene cross-linked pipe (PEX), chlorinated polyvinyl chloride (CPVC), acrylonitrile butadiene styrene (ABS), copper, and galvanized steel. As reported by (Kleczyk, 2012), a survey conducted by Marshutz showed that copper was used in nearly 90% of homes. The second most used was PEX with a 7% installation rate, followed by CPVC with a 2% installation rate. In 2005, a survey conducted via telephone calls with plumbers showed that the use of plastic pipes was increasing due to its lower cost.

The material choice typically depends on the pipe's purpose. For example, copper pipes are commonly used because of their versatility, whereas PEX pipes are used because they are affordable and do not leach rust or metal corrosion products into the water. Galvanized steel is mostly found in older homes (built between 1880-1960) and isn't used in current construction due to its tendency to rust and release trapped pieces of lead in the pipe, degrading the water quality. When modifying and adjusting a plumbing layout, PEX is preferred due to the flexibility of the tubing (Taylor, 2021).

In addition to the piping material, homes and buildings also have solder to connect metal pipes or solvent to connect plastic pipes. Soldering involves heating of a metal alloy at the junction of two pipes, providing a strong bond. Solder is currently made of many different metals such as indium, spelter brass, platinum, gold, and silver. Prior to 1986, solder was often made of lead due to its low melting temperature (Thomas, 2022). However, the corrosion of lead-based solder joints, typically associated with copper service lines, could be a significant source of lead leaching into water (Subramanian & Connor, 1991). Fixtures composed in part of lead may also be a source of lead leaching into water. In 1986, Congress prohibited the use of pipes, solder or flux that are not deemed "lead-free", meaning that solder cannot have more than 0.2% lead, and pipes cannot exceed 8% lead (U.S. Environmental Protection Agency, 2021a).

Several factors contribute to corrosion potential in homes and buildings with metallic components in the premise plumbing system. First, these plumbing systems typically have high surface area to volume ratios which allows for contact of the metal and water. Second, long stagnation times allow for buildup of corrosion products. Lastly, certain water quality conditions can enhance corrosion, as described in Section 1.1.3 (Cullom et al., 2020).

2.1.3 Impact of Chemical Water Quality Parameters on Corrosion

Various chemical water quality parameters can influence corrosion potential, including pH, alkalinity, hardness, chloride and sulfate, dissolved solids, and residual disinfectant. Each of these parameters is described below.

The pH of water is a significant factor in the overall corrosivity of water. Acidic water contains higher levels of H^+ ions, which can react with electrons in the metal to promote galvanic corrosion.

In natural waters, alkalinity is calculated as the sum of the concentrations of bicarbonate (HCO_3^-), carbonate (CO_3^{2-}), and hydroxide equivalents, and is typically reported in units of mg/L as $CaCO_3$. These compounds collectively form a buffer system against changes in pH:

$$HCO_3 + H^+ \rightarrow H_2CO_3 \qquad \text{(Reaction 2-7)}$$

$$HCO_3^- + OH^- \rightarrow CO_3^{2-} + H_2O \qquad \text{(Reaction 2-8)}$$

$$H^+ + OH^- \rightarrow H_2O \qquad \text{(Reaction 2-9)}$$

When acid (H^+) is added to the water, it reacts with the carbonate, bicarbonate, or hydroxide and is neutralized. From a corrosion perspective, waters that are more alkaline or have a higher carbonate concentration are less corrosive, because if waters encounter additional acidity in the distribution system, the pH will not decrease. However, highly alkaline water can also deposit scale on the inside of pipes because alkalinity is related to hardness, as alkalinity is measured in $CaCO_3$, and calcium contributes to hardness. This scale can prevent corrosion by blocking water from reaching the walls of the pipes, but if bacteria become established under the scale, the pipes become vulnerable to microbiologically induced corrosion (Weiner & Matthews, 2003). This has to do with the hardness of the water, which is measured in mg/L of CaCO3. Because the carbonate ion is present, it is also related to alkalinity.

The hardness of water refers to the concentration in mg/L of multivalent cations such as alkaline earth metals. In drinking water, calcium is the most common of these, with some magnesium present and usually only trace amounts of the others. Water high in alkaline earth ions (>200 mg/L) is considered hard, while water low in alkaline earth ions (<100 mg/L) is soft. If the water is undersaturated with $CaCO_3$, it may corrode preexisting protective coatings that are sometimes present in distribution systems. If additional $CaCO_3$ is present above the saturation point, some may precipitate out of the water, depositing scale onto pipe walls, appliances, or other metal surfaces such as sinks. This quality, along with a difference in taste, can make hard water unpopular with consumers (Droste & Gehr, 2018).

Chlorides and sulfates can also cause corrosion of lead when present in drinking water systems. Ng and Lin (2015) conducted experiments changing pH, chloride, and sulfate concentrations in drinking water over a 7-day period. According to their results, the concentration of lead leached into the water increased with increased amounts of chlorides and sulfates. Chlorides and sulfates increase electrolyte conductivity, and high conductivity encourages corrosion across the surface rather than concentrated in one spot, increasing galvanic corrosion (Ng & Lin, 2015).

Total dissolved solids (TDS) are all inorganic and organic substances that are dissolved in water. They are small enough to fit through a 2-micrometer sieve. They are usually constituted of salts and metals such as calcium, magnesium, carbonates, sodium, chloride, and sulfate. TDS comes from a variety of sources such as sewage, runoff from urban areas and agricultural practices, and deicing salts. Waters with high total dissolved solids levels (>500 mg/L) are often seen as having higher corrosion potential because of ions such as chloride (World Health Organization, 1996). When water has low total dissolved solids levels (<50 mg/L), it is also common for the water to have corrosive tendencies. This is because the water likely has low alkalinity levels and therefore cannot buffer pH changes (Master Water Conditioning Corp, 2019).

Chloramine (NH_2Cl) is a disinfectant often used in drinking water systems as a secondary disinfectant in the distribution system. However, chloramine can affect the corrosion of lead and copper. Chloramine can change the chemical properties of water which can impact pH and alkalinity, which as stated above, can increase the likelihood of corrosion from pipes and fittings. The addition of chloramine can also result in nitrification if not correctly utilized. Nitrification occurs when organic matter or bacteria oxidizes ammonia (NH_4^+) to nitrite (NO_2^-) and then to nitrate (NO_3^-). The formation of the nitrate ion will decrease the pH of the water and therefore increase the likelihood of corrosion (U.S. Environmental Protection Agency, 2022a).

2.1.4 Impact of Physical Water Quality Properties on Corrosion

Temperature and flow are two physical properties that affect corrosion. As temperature and flow change there are direct correlations to corrosion rates, these will be explored more below.

Temperature plays a large part in corrosion. The higher the temperature the faster electrochemical reactions occur. The higher the temperature, the more energy is added into the system which results in an increase in corrosion rates. Generally, for an increase of 10 degrees Celsius the corrosion rate doubles.

Another physical property that affects corrosion is flow. If there is no flow, the water is described as stagnant. Stagnant water leads to the growth of microorganisms, which can create a biofilm which is a complex microbiome that adheres to the pipe surface. The biofilm composition can vary depending on oxygen concentrations, nutrient concentrations, and pH conditions within the film. When there is biofilm over an area in a pipe, the area underneath experiences corrosion due to the biofilm and its interactions with its environment and metal, the biofilm then corrodes by altering concentration of ions, pH value, and oxidation reduction potential (Telegdi et al., 2017).

Corrosion can also be increased due to high water velocities. The velocity of a fluid and the effect of a fluid moving along a pipe surface combine with the corrosive action of the fluid moving. These actions result in an increase in the loss of metal. Normally the metal is protected with a film, but if erodes then corrosion begins on the bare surface of the metal. Higher water velocities can increase corrosion as it moves more water along the surface which only results in losing more metal and worsening corrosion.

2.2 Adverse Effects of Lead and Copper in Drinking Water

Lead and copper in drinking water caused by corrosion can have several negative effects. First, any damage to pipes from corrosion has the potential to reduce the lifetime of the infrastructure and cause problems sooner than they might otherwise occur. Excess lead and copper in water can also cause unpleasant taste, odor, and aesthetics. In addition, both lead and copper are associated with health problems at levels that could be seen in drinking water.

2.2.1 Pipe Damage

As pipes corrode internally, they become less thick and are more prone to leaks or bursts. As described in Section 1.1.3, pipes are at an increased risk of corrosion at joints between two different metals. This means that typically, these joints will be the site of any leaks that occur. Leaky pipes need to be repaired or replaced, which can be expensive and difficult. If they are not replaced in a timely manner, they will continue to waste water, leading to a higher water bill for the resident/customer and potential water damage to surfaces in the area surrounding the pipe. A large enough leak or burst can create a drop in pressure sufficient to cause service issues downstream from the problem.

2.2.2 Aesthetic Effects

The presence of lead and copper in water can cause aesthetic effects. Metals in water are not typically known to have a taste, but rather a “mouthfeel”, which is a feeling in the mouth due to a sensation of oral nerve endings. Depending on each individual's taste portfolio, copper has been said to produce unpleasant sensations such as acidic, salty, bitter, or astringent (Deitrich et al., 2004). The metallic, unpleasant taste that copper causes in water typically occurs when copper is at high levels of concentration in the water (Government of Western Australia Department of Health, 2016). In addition to taste, the presence of copper in water can cause laundry and plumbing fixtures to become stained in color. A low level of copper (1 mg/L) in water creates a blueish-green stain on certain fixtures such as taps, pipes, showers, toilets, and hand basins, as shown in Figure 2 (Government of Western Australia Department of Health, 2016).

Figure 2. Sink Stained from Copper in Water (Mr. Plumber, n.d.)

2.2.3 Health Effects of Lead

Ideally, drinking water distribution systems and premise plumbing would be made of materials that will not cause any harm to consumers. However, when a drinking water system contains plumbing materials with lead, this lead can corrode and end up in the drinking water (U.S. Environmental Protection Agency, 2022c). Lead in drinking water can cause negative human health effects, especially in children and pregnant women. Because of potential health impacts, the maximum contaminant level goal of lead is set at zero.

Children, especially those 6 years and younger, are among the most vulnerable when it comes to lead exposure. There is no safe amount of lead for the human body. According to the Mayo Clinic (2022), lead poisoning can be extremely hard to detect in the human body until it is already at elevated levels in the blood, and in children, this is around 5 µg/dL. Signs and symptoms of lead poisoning include developmental delays, learning difficulties, irritability, loss of appetite, weight loss, sluggishness, fatigue, abdominal pain, vomiting, constipation, hearing loss, seizures, and pica, an eating disorder in which the affected person eats things that are not normally considered food. At extremely elevated levels, lead poisoning can be fatal, which is 10 µg/dL (Needleman & Landrigan, 2004). It has been shown that any amount more than 0 µg/dL of lead in the blood can cause damage to the central and peripheral nervous system, behavioral and learning disabilities including lower IQ and hyperactivity, stunted growth, impaired hearing, and impaired formation and function of blood cells including anemia (U.S. Environmental Protection Agency, 2022c).

Ingestion of lead is also of concern for newborns and pregnant women. According to the National Center for Environmental Health: Division of Emergency and Environmental Health Services (2010), there have been a few studies showing that lead exposure as low as 3 µg/dL can result in delayed sexual maturation and a reduction in infertility. Lead exposure can lead to hypertension, also known as high blood pressure, which is a common complication during pregnancy. Lead can damage the vascular endothelium, which is the cellular lining of arteries, veins, and capillaries that controls blood pressure. Damage to the endothelium can cause hypertension and preeclampsia, a pregnancy complication resulting in high blood pressure and damage to other organs, usually the liver and kidneys. At high blood exposure (≥30 µg/dL), spontaneous abortion or miscarriage, the loss of the fetus naturally before 20 weeks, is extremely common. Lead exposure in pregnant women is also linked with complications with the newborn such as premature delivery, reduced growth, and lower birth weights.

While children, newborns, and pregnant women are at the highest risk for severe health effects due to lead exposure, lead can cause negative health impacts to the adult human body as well. At high enough levels, those about 80 µg/dL, lead exposure in adults can cause cardiovascular impacts, high blood pressure/hypertension, decreased kidney function, and reproductive issues (U.S. Environmental Protection Agency, 2022c). Lastly, it is imperative to know that once lead gets into the bloodstream, it is incredibly difficult to remove.

2.2.4 Health Effects of Copper

Copper is commonly used for water pipes in homes, and therefore can be introduced into drinking water through corrosion. There are also low levels of copper present in all water sources, about 0.01 mg/L in surface water from copper being a natural and commonly occurring metal, while copper in pipe systems is most likely from copper pipes and plumbing found in homes. Consuming copper in the diet is necessary for good health as copper is an essential nutrient to the human body. The US Department of Agriculture recommends 0.9 mg a day for most adults.

While copper is an essential nutrient, high concentrations can cause negative health effects (Government of Western Australia Department of Health, 2016). The EPA action level for copper is 1.3 milligrams/liter (mg/L) for the 90th percentile value, which means that more than 10% of water samples have a value equal to or above 1.3 mg/L (U.S. Environmental Protection Agency, 2021b). Consumption of excess copper can lead to nausea, vomiting, diarrhea, and stomach cramps. Copper exposure of 3 mg/L or greater is considered very high and could result in kidney and liver damage which could result in death (Agency for Toxic Substances and Disease Registry, 2011).

Exposure to copper at high levels is believed to cause the same effects in children as it does in adults. It is not known if the effects occur at the same dosage level as adults, or if children experience more severe effects. High copper levels are also not known to cause any birth defects or developmental effects (Centers for Disease Control and Prevention, 2013).

There is a genetic disease which increases the severity of health effects due to excess copper exposure. Wilson disease is where the body has trouble eliminating copper, resulting in excessive accumulation of copper in the brain, liver, and eyes. This often leads to liver disease and can also result in nervous system or psychiatric problems. Treatment involves lifelong commitment to low-copper diet and medication (National Institute of Health, 2020).

2.3 Regulations

The Safe Water Drinking Act (SDWA) was established in 1974 and then revised in 1986 and 1996, to protect the public and maintain safe standards for public drinking waters (U.S. Environmental Protection Agency, 2004). The SDWA requires the Environmental Protection Agency (EPA) to set limits for contaminants that can have adverse health effects. First, the EPA determines levels at which contaminants in drinking water would have no adverse health effects. These levels are known as maximum contaminant level goals (MCLGs) and are non-enforceable. Next, the EPA sets an enforceable standard that is as close to the MCLG as possible, taking into consideration technical and economic feasibility. Depending on the contaminant, the enforceable standard is a maximum contaminant level (MCL), a treatment technique (TT), or action level (AL). A maximum contaminant level is the highest level a contaminant can be in drinking water and is often set as close to the MCLG as possible (U.S. Environmental Protection Agency, 2022e). The key difference between an MCL and MCLG is that an MCL is an enforceable standard. A treatment technique is put in place as an attempt to lower the contaminant levels in drinking water. An action level is a level at which provisions laid out in the rule must be initiated by the water system (Centers for Disease Control and Prevention, 2017). As described in the following sections, the EPA has set the MCLG for lead at zero, due to lead's toxicity and bioaccumulation characteristics, and the MCLG for copper at 1.3 mg/L due to the possibility for liver and kidney damage at high exposure (U.S. Environmental Protection Agency, 2022c). In addition to the MCLGs, the Lead and Copper Rule created an action level of 0.015 mg/L for lead and 1.3 mg/L for copper (U.S. Environmental Protection Agency, 2008).

2.3.1 Lead and Copper Rule (1991)

Lead and copper are most likely to enter drinking water systems through corrosion of plumbing materials containing lead and copper. In 1991, the EPA created a regulation to control the amount of lead and copper in drinking water to protect the public's health (U.S. Environmental Protection Agency, 2021b). This regulation is the Lead and Copper Rule (LCR). This regulation applies to community water systems as well as non-transient, non-community water systems, referred to throughout the rule as "water systems". The rule replaced the previous standard of 50 parts per billion (ppb) of lead at the entry point of the distribution system.

The LCR set the action limit for lead at 0.15 mg/L (15 ppb) and for copper at 1.3 mg/L (1300 ppb) for 90th percentile values. If a water is in exceedance of either action limit, the water utility must take action to reduce corrosion in the water system. The 90th percentile value is the value for which 90% of samples are at or below the value. To calculate the 90th percentile value, lead (or copper) values are placed in ascending order. Then, the number of samples is multiplied by 0.9. The 90th percentile value is the concentration that corresponds to the resulting number. For example, if a utility collects10 samples, the concentration of the 9th sample would be the 90th percentile (because $10 \cdot 0.9=9$), and if a utility collects 50 samples, the concentration of the 45th sample would be the 90th percentile value ($50 \cdot 0.9=45$ $50 \cdot 0.9=45$). If a utility collects 5 samples, the resulting number is 4.5 and therefore the utility would average the concentrations of the 4th and 5th samples to determine a 90th percentile value. By using the 90th percentile, the LCR allows for outliers in sampling. The provisions are intended to reduce corrosion and thus reduce concentrations of lead and copper at consumer taps, and these provisions include water quality parameter monitoring, corrosion control treatment, source water monitoring and treatment, public education, and lead service line replacement (U.S. Environmental Protection Agency, 2008). The LCR created a maximum contaminant level goal (MCLG) of zero for lead and 1.3 mg/L for copper in drinking water and provided information on treatment techniques to reduce the corrosion of lead and copper in distribution systems.

According to the LCR, all community water systems, and non-transient, noncommunity water systems must perform first-draw testing in homes that are at high risk of lead and copper contamination. High-risk homes are determined based on the age of the home, the presence of a lead service line, and previous lead contamination in the area. The first-draw test is conducted on water that has stagnated in the plumbing for at least 8 but no more than 18 hours and is usually conducted on the water in the tap first thing in the morning, hence the name "first-draw" (Oregon Department of Human Services, n.d.). The water system provides selected homeowners with sampling containers. The homeowners fill the containers with first-draw samples of water from the kitchen in the morning and return the samples to the water system for testing. In addition to testing for lead and copper concentrations, water quality parameter monitoring, where water is sampled and analyzed for pollutants, pesticides, metals, and oils, is performed on water systems that serve over 50,000 people, or those that serve less than 50,000 but have exceeded the action limit set by the LCR (U.S. Environmental Protection Agency, 2012). Samples are collected from consumers' taps and at the distribution system entry points every six months. If corrosion control treatment has been initiated/changed, samples are collected at the distribution system entry points every two weeks.

The number of samples collected from residences is dependent on the system size and can be seen in Table 2. The standard sampling protocol includes sampling every six months, with the "standard" number of samples. Both the frequency of sampling and number of samples can be reduced if a system meets certain criteria (U.S. Environmental Protection Agency, 2008). A system can qualify for annual testing and a reduced number of samples if (1) the water system serves less than 50,000 people and sampling results are less than or equal to both action limits for two consecutive six-month periods or (2) the water system meets optimal water quality parameters and is less than or equal to the lead action limit for two consecutive six-month periods. There are three ways to qualify for triannual testing with reduced number of samples: (1) the water system serves less than or equal to 50,000 people and has sampling results that are less than or equal to both action limits for 3 consecutive years of monitoring; (2) the system meets optimal water quality parameter specifications and has sampling results less than or equal to lead action limit for three consecutive years of monitoring; or (3) any system with 90th percentile lead and copper levels less than or equal to 0.005 mg/L and less than or equal to 0.65 mg/L, respectively, for two consecutive six-month periods. Lastly, for a system to qualify for testing every nine years, the system must serve less than or equal to 3,300 people and meet monitoring waiver criteria.

Table 2. Number of LCR Samples Collected Based on System Size (U.S. Environmental Protection Agency, 2008)

Size Category	System Size	Number of Pb/Cu Tap Sample Sites		Number of WQP Tap Sample Sites	
		Standard	Reduced	Standard	Reduced
Large	> 100K	100	50	25	10
	50,001 - 100K	60	30	10	7
Medium	10,001 - 50K	60	30	10	7
	3,301 – 10K	40	20	3	3
Small	501 – 3,300	20	10	2	2
	101 - 500	10	5	1	1
	<100	5	5	1	1

Since the Lead and Copper Rule was created, the number of drinking water systems in the nation with concentrations exceeding the action levels has decreased by over 90%. By implementing the LCR, the risk of becoming exposed to lead and copper has significantly decreased, thereby reducing the probability of detrimental health effects.

2.3.2 Minor Revisions to the LCR (1999 – 2007)

Since 1991, the Lead and Copper Rule has been revised many times, adding and modifying aspects of the original Rule (U.S. Environmental Protection Agency, 2021b). In 1999, the EPA made minor changes to the LCR known as Lead and Copper Rule Minor Revisions (LCRMR). These revisions did not change the action levels for lead or copper, and also did not alter the Maximum Contaminant Level Goals. The changes to the LCR had seven broad categories. The first category was the Demonstration on Optimal Corrosion Control, which involved adding a requirement for systems with little to no corrosion to monitor every 3 years, changed the way compliance with the state water quality parameters was determined, and clarified some requirements and wording already in place. The second category was Lead Service Line Replacement Requirements which required water system facilities to identify and replace lead service lines within the distribution system areas that they own and to notify the residents of the areas with these service lines. Public Education Requirements was the third category, with changes in how information about lead and copper can be provided to the public. The fourth category was Monitoring Requirements, which had the most revisions. Some of these revisions were allowing more flexibility for the time of year systems can reduce monitoring, allowing states to waive testing to small systems with plumbing without lead and copper materials, clarifications and flexibility in sample site requirements, and added a requirement for systems to report changed in treatment to the State. The fifth category was Analytical Methods that contained one update to the analytical methods of lead and copper to make them conform more with the changes in the methods for other inorganic chemicals. The sixth category was Reporting and Record Keeping Requirements. The revisions to this section streamlined state reporting requirements, removing system reporting requirements that were no longer needed. The final category is Special Primacy Considerations which added considerations when multiple samples are collected per day. These revisions took effect on April 11, 2000 (U.S. Environmental Protection Agency, 1999).

In 2004, more revisions were made to the Lead and Copper Rule. These revisions were mostly textual, including adding information back into the rule that had been accidentally dropped. This information listed the facilities that had to be sent public education brochures for public water systems that had lead and copper over the allowed limit. The revisions also fixed typographical errors, editing errors, and updated language and terminology. Finally, these revisions added a section on optional monitoring for disinfection profiling as well as a detection limit for Uranium Methods. These revisions took effect on July 29, 2004 (U.S. Environmental Protection Agency, 2021b).

In 2007, the EPA made “Short-Term Revisions to the Lead and Copper Rule”. These revisions aimed to enhance monitoring, customer awareness, treatment, and lead service line replacement. Additionally, the revisions aimed to improve the public education requirements by ensuring that drinking water consumers get timely, meaningful information to inform them of contaminated waters and how to limit their exposure to lead in their drinking water (U.S. Environmental Protection Agency, 2021b).

2.3.3 Revised Lead and Copper Rule (2021)

On June 10, 2021, the EPA signed a rule to extend the effective date of these new revisions to December 16th, 2021, in order to allow the EPA to continue to gather input from vulnerable communities. These revisions ultimately aim to better protect children and communities through better detection of lead and protection of consumers from lead in water at schools and childcare facilities. One way of doing this is continuing programs to replace lead service lines. Other additions to the rule are to use science-based testing protocols to identify sources of lead, and to establish a trigger level that will call attention to the source and start mitigation sooner. Lastly, the revised rule requires testing in schools and childcare facilities. This is the first time this has been included in the Lead and Copper Rule and will provide a more complete understanding of the composition of the waterways entering the schools and their materials, enabling water systems to start mitigation sooner. This revision developed a new regulation called the Lead and Copper Rule Improvements (LCRI) to accomplish these goals and enhance the measures in place to protect communities from lead in drinking water. (U.S. Environmental Protection Agency, 2021c).

2.4 Predicting Corrosion Potential

When designing water treatment plans or addressing existing problems, water quality engineers need ways to predict how corrosive a water will be to piping in the distribution system. There are several indices that can be calculated using water quality data and may predict corrosion potential for certain piping materials. Rates of corrosion can be directly measured in pipes using either the pipe loop method, which involves running water through a pipe for a long period of time, or using a coupon rack, where metal strips are exposed to flowing water to simulate pipe conditions.

2.4.1 Corrosion Indexes

Corrosivity can be predicted through different corrosivity indexes. There are three indexes that are commonly used, which are Langelier Saturation Index (LSI), Potential to Promote Galvanic Corrosion (PPGC), and the Larson Ratio (LR). Each index calculates the corrosion potential of the water based on different information (Water Resources, 2019).

The LSI is the measure of the potential for $CaCO_3$ precipitation. The index ranges from –2.0 to 2.0. The pH, temperature, total dissolved solids, alkalinity, and total hardness are used to find how saturated water is with calcium carbonate. A negative LSI indicates the water is under-saturated with calcium carbonate. When under-saturated the water has a greater tendency to remove calcium carbonate from protective coatings. A positive LSI indicates over saturation of calcium carbonate which increases the likelihood that scaling will form and protect the pipe surface.

The PPGC uses the ratio of chloride to sulfate and is also known as The Chloride to Sulfate Mass Ratio (CSMR). This index ranges from below 0.2 to greater than 0.5, with the greatest corrosion potential being when greater than 0.5 while having less than 50 alkalinity present. The higher the CSMR, the higher the PPGC ratio, which indicates a greater presence of dissolved chloride. Dissolved chloride increases the solubility of lead resulting in a greater potential for corrosion on galvanic lead. The lower the CSMR the lower the PPGC ratio, which indicates an elevated presence of sulfates. A greater presence of sulfates results in a lower corrosion potential as the sulfates would form a lead-sulfate precipitate.

The LR uses the sum of equivalents for chloride and sulfate, divided by the equivalents of bicarbonate. The higher the ratio, the more likely corrosion will occur. The LR is used to describe the corrosivity of water towards iron and steel.

2.4.2 Pipe Loop Testing

Pipe loop testing is a test for corrosion. Pipe loop tests are typically conducted in a laboratory setting with a section or multiple sections of pipe isolated. The pipes can either be new or cut from an existing system. The pipes are made into a loop and water representative of the distribution system water is then run through each section constantly. This type of testing allows for various factors to be controlled to see their impact on the pipe and quality of the water. Different pipe materials can be tested to see how the same water and flow conditions will affect different materials. The water quality and flow characteristics can also be changed to see how these factors would impact corrosion in the pipe, or to see how effective possible corrosion treatment would be.

While this test might allow for variables to be controlled to see the impact of changes, this test usually takes a minimum of six months, but is often run for years. Because of the length of testing time, these tests are costly to conduct (Civardi & Gray, 2005). Due to the time and cost, narrowing down the variables or factors being tested is important.

2.4.3 Coupon Testing

Coupon testing is a test to indicate corrosion in a system. Coupons are metal strips that are pre-weighed and are selected to represent the metals that are present in a particular piping system. The coupons are mounted inside a coupon rack, which is a length of pipe mounted in a back-and-forth pattern on a solid backboard and connected to a water system. The piping in a coupon rack is typically made of PVC or a similar material so no corrosion occurs from the pipe. The metal strips are typically exposed to water flowing through the coupon rack for 60, 90, or 120 days. After removal from the coupon rack, the metal strips have their initial weight compared to the final weight, and the difference in weight is used to estimate the rate of corrosion in units of Mils penetration per year (MPY), where a Mil is equal to one thousandth of an inch. This testing method only reflects the conditions at the site of the coupon rack, resulting in temperature having little effect on results, when temperature plays a large part in corrosion (Welsh, 2021).

2.5 Corrosion Control Methods

Corrosion can damage the interior of pipes and result in harmful chemicals getting into the water. There are counter measures that can be taken to address corrosion. System-wide, utilities can adjust water quality or introduce corrosion inhibitors. On the local scale, replacement of individual lead service lines is another form of corrosion control. These controls are discussed in the following sections.

2.5.1 pH Adjustment

One of the primary methods that water systems use to reduce the corrosivity of their water is raising the pH. As discussed in Section 1.1.3, low pH causes water to be more corrosive because it contains more H^+ ions in solution that increase the prevalence of galvanic corrosion by reacting with electrons in the pipe. While other water quality parameters affect corrosion, pH is one of the most impactful parameters and the easiest to alter, making it a common target for treatment plants that want to reduce their corrosivity.

To control lead and copper corrosion of plumbing, the target pH for a water should be 8.8 to 10. If a system is not using a corrosion inhibitor, the pH should be further increased to 9.0 or greater. Lower pH values likely result in a poor buffering quality of the water, making it more likely to corrode the plumbing. pH is related to alkalinity and dissolved inorganic carbon (DIC). For example, the higher in the 8.8-10 pH range that the water is, the less DIC is needed to buffer the water. This is crucial to monitor, since the higher the DIC concentration (above 20 mg/L as calcium), the more lead is released into the water (U.S. Environmental Protection Agency, 2016).

2.5.2 Corrosion Inhibitors

Corrosion inhibitors are chemicals that decrease the corrosion rate in a system without changing the concentration of any corrosion agent (de Damborenea et al., 2014). Corrosion inhibitors can usually be effective in small concentration, unless it is a chemical aimed at substantially reducing pH, or oxygen and hydrogen sulfide, which causes the removal of aggressive species and likely would need a higher concentration. Corrosion inhibitors can be cathodic, anodic, or mixed. This classification depends on if their influence is mainly in delaying the cathodic reaction, the anodic reaction, or both the cathodic and anodic reaction of the corrosion process. These inhibitors cause the corrosion potential to shift in the desired cathodic or anodic direction or leave the metal corrosive potential unchanged.

The most common corrosion inhibitors are phosphate-based with orthophosphate (PO_4^{3-}) in their formula (U.S. Environmental Protection Agency, 2016). Occasionally, silicate-based inhibitors are used, which is a mixture of silicon dioxide and soda ash. Orthophosphate is most commonly used to control lead and copper corrosion. Polyphosphates can seclude and sequester lead and copper, which increases the risk of exposure, making them generally not effective for controlling lead and copper release into the water system. However, blended phosphates, which is a mixture of orthophosphates and polyphosphates, have been used to sequester manganese and iron and control corrosion. Additionally, silicate-based inhibitors reduce the lead and copper levels of first draw samples, however, have a limited full-scale success.

Orthophosphate reacts with divalent copper and lead (Pb^{2+} and Cu^{2+}) to create compounds that tend to stay in solid form. Orthophosphates are anodic corrosion inhibitors. The strength of these bonds and of orthophosphates' ability to control lead and copper release into the water is dependent on the orthophosphate concentration, pH, DIC, and characteristics of the corrosion scale (if the water contains other metals). Orthophosphate comes in many forms: phosphoric acid, salt (potassium or sodium), and zinc orthophosphate. Zinc orthophosphate inhibitors usually have a 1:1 ratio of zinc: phosphate. Research shows that zinc orthophosphate inhibitors do not display higher control than orthophosphate, however the zinc provides better corrosion protection for cement that is at low alkalinity/pH/hardness. Phosphoric acid (H_3PO_4) is a commonly used form that is available in concentrations between 36-85% (U.S. Environmental Protection Agency, 2016).

Blended phosphates have been proven to be effective in reducing lead levels, however they are more vulnerable to physical disturbances and low water use. Additionally, it is unknown whether blended phosphates can effectively control copper corrosion, specifically at high alkalinities. Calcium and aluminum greatly affect the effectiveness of blended phosphates since blended phosphates create a barrier film from the interactions of calcium and aluminum in the water with phosphorous containing compounds. If calcium and aluminum are not present or are only present in low concentrations, the effectiveness of blended phosphates is hindered. The EPA recommends a demonstration study followed by extensive monitoring for systems that choose to use blended phosphates (U.S. Environmental Protection Agency, 2016).

Silicate inhibitors are available in liquid or solid form. While silicate inhibitors have shown to reduce lead and copper levels in first draws, they have not been used in many full-scale plants. Silicate inhibitors are typically more expensive than phosphate-based inhibitors and typically require high doses, therefore increasing the cost. Silicates increase the pH of water, which may reduce the release of lead and copper. Additionally, silicate may form a film on the pipes surface that acts as a diffusion barrier. However, the effectiveness of this diffusion barrier depends on the pre-existing corrosion products (U.S. Environmental Protection Agency, 2016).

2.5.3 Lead Service Line Replacement

Lead service lines are the most common source of lead and copper contamination in drinking water, and it is estimated that there are somewhere between 6 and 10 million lead service lines in the US (U.S. Environmental Protection Agency, 2022d). Because of this, a form of corrosion control is replacing lead service lines and is considered the only long-term solution to protecting the public from lead contamination, although there is no 100% effective measure that can be taken.

In lead service line replacement, the lead service lines are removed and replaced with a safer alternative such as copper lines. However, there are many challenges when it comes to completing LSLR, one being the fact that there are both public and privately-owned lead service lines. When replacing these lines, it is imperative to replace the entire line, as replacing only part of it has actually been seen to increase lead levels in the water (Environmental Defense Fund, 2022). This means that when LSLR is completed, the entire line needs to be replaced, which could be both partially and publicly owned, which can cause a lot of bureaucratic issues and increase the period of replacement. It also begs the question of who is responsible for paying for the replacement.

LSLR is also quite expensive and destructive for private landowners and public communities to deal with, so the EPA has provided information for potential funding sources and other resources available to those in need of lead service line replacement (LSLR) to help manage the cost. The Drinking Water State Revolving Fund (DWSRF), the Water Infrastructure Improvements for the Nation Act (WIIN) Grants, the Water Infrastructure Finance and Innovation Act (WIFIA), and the HUD Development Community Block Grant Program all have federal funding available for LSLR (U.S. Environmental Protection Agency, 2022d). Communities and individuals may apply for these funding sources to help offset the cost of LSLR. On average, to replace one-line costs about $4,700 but can range anywhere from $1,200 to $12,300 per line. In addition to the cost and ownership complications that come with LSLR, a lot of water utilities do not have exact data on how many and where all of the lead service lines are located, which makes it exceedingly difficult to determine what needs to be replaced (Environmental Defense Fund, 2022).

There are nine communities across the United States that have taken action and have completely removed all lead service lines from their communities and should be used as a model for other communities still struggling to complete this process. A few of these communities include Framingham, Massachusetts, which completed its replacement in April of 2016 after laying out a comprehensive plan in 2005. Other successful communities include Green Bay, Wisconsin completed in October of 2020, Lansing, Michigan completed in December of 2016, and Madison, Wisconsin completed in 2011, among a few others. These communities have shown that while it may be expensive and time-consuming, it is a possible and practical effort to be made.

3. Methods

Current methods to assess the corrosion potential of drinking water take months to years to complete. The goal of this project was to determine whether a short-term laboratory experiment would provide useful data to predict corrosion potential. To achieve this goal, samples of water with varying pH, alkalinity, and temperature were created and lead coupons were added to each sample. The dissolved lead concentrations were measured after 1 day, 1 week, 3 weeks, and 4 weeks. This chapter presents the experimental design and analytical methods for the experiments.

3.1 Experimental Design

Experiments were conducted to monitor the corrosion of lead coupons in waters with different properties. First, waters were created with varying pH and alkalinity. Then, lead coupons were added to each sample and incubated at either 4°C or 20°C. At various time intervals, aliquots were removed from each water sample for dissolved lead testing. At the conclusion of the testing period, dissolved lead, pH, alkalinity, and lead coupon weight were measured for each test. Figure 1 shows the overall experimental design, and details of these tests are provided in the following sections.

3.1.1 Water Samples

As shown in Figure 3, the corrosion of lead coupons was tested in 20 samples. Ten samples were used to assess the impact of alkalinity on corrosion. Alkalinity was adjusted by the addition of sodium bicarbonate (see Section 3.2.4) to values of 10, 30, 75, 100, and 200 mg/L as $CaCO_3$. The 10 mg/L samples represented low alkalinity waters, while the remaining values represented the range of alkalinities found in typical drinking waters. The pH in these samples was allowed to vary slightly as alkalinity was the primary focus, however, the variation was kept to within 1 pH. Two samples were created at each alkalinity with one incubated at 4°C and one incubated at 20°C for a total of 10 alkalinity samples.

The remaining ten samples were used to assess the impact of pH on corrosion. pH was adjusted by the addition of 0.1N HCl or 0.1N NaOH (see Section 3.2.3). The pH goals were 3.0, 6.5, 7.5, 8.0, and 12.0. The lowest value of 3 represented a strong acid. Values of 6.5, 7.5, and 8.0 are within the range recommended by the EPA for drinking water distribution systems to control corrosion and disinfection byproducts. The final pH of 12 represents a strong base. As with the alkalinity samples, two samples were created at each pH condition, with one stored at 4°C and one stored at 20°C. To help keep pH stable, a phosphate buffer was used for the 6.5, 7.5, and 8.0 samples.

After the solutions were made, one coupon was inserted into each water sample, and the samples were placed in incubators. The samples were removed at the predetermined time intervals (1 day, 1 week, 2 weeks, and 4 weeks), and 10 mL aliquots were removed from each beaker and stored in polypropylene tubes. The beakers were returned to the appropriate incubator/refrigerator, and the tubes were stored at 4°C until tested for dissolved lead concentrations. After 4 weeks, all samples were removed from the incubator/refrigerator and analyzed for pH, and samples with added alkalinity were tested for alkalinity concentration.

3.1.2 Temperature

Two temperatures were used for the experiments: 4°C and 20°C. The low temperature was selected as corrosion potential is lower at lower temperatures, while the high temperature was selected to increase the corrosion potential. The high temperature was achieved by placing samples in a Precision 815 incubator set to 20°C.

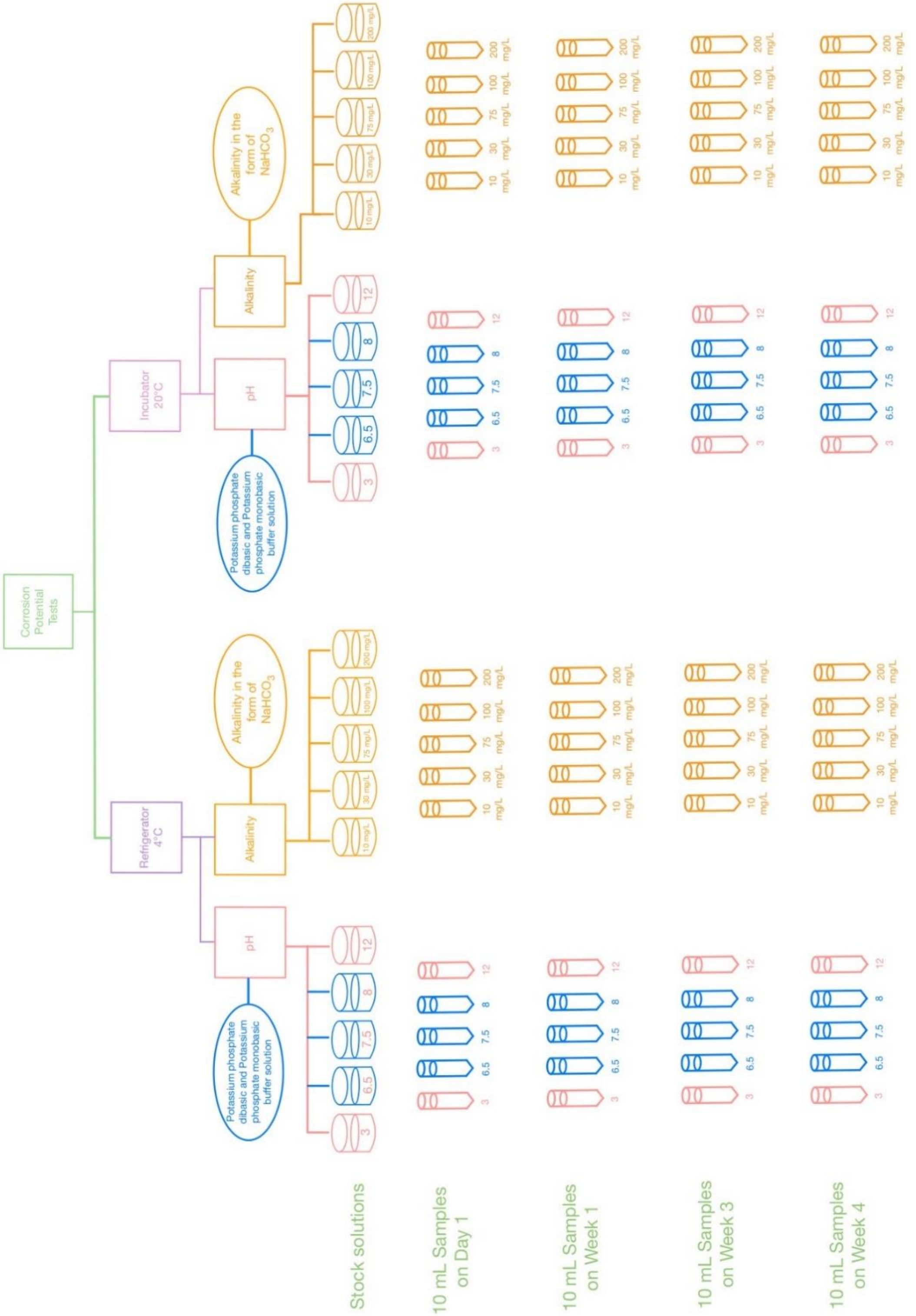

Figure 3. The Experimental Structure

The low temperature was achieved by placing samples in a Wood’s frost-free refrigerator set to 4°C. To verify the temperatures, laboratory thermometers were placed in both the incubator and refrigerator.

3.1.3 Lead Coupons

Lead coupons were purchased from Pacific Sensor (Carrollton, Texas). The coupons were made of grade A lead alloy (UNS L50050), were mill finished, and were approximately 3” x ½” x 1/16” in size. Each coupon had a serial number and was pre-weighed with a certificate of the weight in grams. Prior to the experiments, the weights were verified and recorded using an analytical balance. At the conclusion of the experiments, the coupons were air-dried and re-weighed.

3.1.4 Time Intervals

The common tests to determine corrosion potential based on lead dissolution are coupon rack tests which take 2-3 months; and pipe loop tests which take 12 or more months. Therefore, the bench-scale tests were designed to take days to weeks. The intent was to shorten the time for a utility to have corrosion potential data while allowing sufficient time for corrosion to occur. The tests were conducted for four weeks, with samples collected for dissolved lead at one day, one week, three weeks, and four weeks.

3.2 Analytical Methods

This section provides details on methods used to measure weight, pH, alkalinity, and dissolved lead concentrations, and methods used to create the water samples.

3.2.1 Weight

A Mettler Toledo MS104TS/00 analytical balance was used for measuring lead coupon weights and chemicals. For all measurements, a weighing dish was placed on the balance and the balance tared with the doors closed. Then, the item to be weighed was added to the dish, the doors were closed, and the weight was recorded when a stable reading was achieved.

3.2.2 pH Measurements

An Accumet basic AB15 pH meter was used for pH measurements. The pH meter was standardized with 4, 7 and 10 pH buffers on each day of use. To measure pH in samples, the probe was rinsed with deionized water, placed in the sample, and the pH was recorded when a stable reading was achieved.

3.2.3 pH Adjustment

As mentioned in Section 3.1, five pH conditions were tested: 3.0, 6.5, 7.5, 8.0, and 12.0. The high and low pH solutions were created by addition of 0.1N HCl or 0.1N NaOH to distilled water. The mid-pH solutions were created using a phosphate buffer solution and 0.1N HCl or 0.1N NaOH, as maintaining mid-pH values in distilled water was not possible without a buffer.

First, a 0.1N hydrochloric acid (HCl) solution was created as a volumetric dilution of a 10N HCl solution. The following equation was used:

$$N_1V_1 = N_2V_2 \qquad \text{(Equation 3-1)}$$

In Equation 3-1, N and V are the normality and volume, respectively, 1 refers to the 10N stock solution and 2 refers to the 0.1N solution. Using Equation 3-1, it was determined that combining 0.98 mL of 10N HCl with 99.02 mL of distilled water would create 100 mL of 0.1N HCl. To create the sample for experimentation, a transfer pipette was used to add 0.1N HCl to distilled water until the desired pH of 3.0 was reached.

A similar process was followed to prepare the pH 12.0 solution, using NaOH instead of HCl. First, a 0.1N NaOH solution was prepared. Then, the 0.1N NaOH was added to distilled water until the desired pH of 12 was reached.

To create the solutions with mid-range pH conditions, potassium phosphate dibasic (K_2HPO_4) and potassium phosphate monobasic (KH_2PO_4) were used in varying amounts as these chemicals create a buffer solution that can maintain pH. First, the 6.5 pH solution was made. 5.9 grams of potassium phosphate dibasic and 9.0 grams of potassium phosphate monobasic were measured. These weighed solutions were then added to distilled water, and then the water was increased to 1L volume. Using HCl, the solution was adjusted to the desired pH.

To create the 7.5 pH solution, 12.8 grams of potassium phosphate dibasic and 3.6 grams of potassium phosphate monobasic were dissolved in a total volume of 1L, and the pH was adjusted using NaOH. Lastly, to create the 8.0 pH, 16.3 grams of potassium phosphate dibasic and 0.888 grams of potassium phosphate monobasic were dissolved up to a total volume of 1L, and NaOH was used to adjust the pH to the desired value.

3.2.4 Preparation of Solutions with Varying Alkalinity

Sodium bicarbonate ($NaHCO_3$) was used to vary alkalinity in the samples. The amount of $NaHCO_3$ required to create the solutions was determined by converting mg/L as $CaCO_3$ to g/L of $NaHCO_3$. An example calculation for the 100 mg/L as $CaCO_3$ solutions is shown in the following calculation:

$$100\ \frac{mg}{L} CaCO_3 \times 2\ \frac{eq}{mol} \times \frac{1}{100} \frac{mol}{g} \times \frac{1\,g}{1000\,mg} = 0.002\ \frac{eq}{L} \qquad \text{(Equation 3-2)}$$

$$0.002\ \frac{eq}{L} \times\ 1 \frac{mol}{eq}\ NaHCO_3 \times 84\ \frac{g}{mol} NaHCO_3 = 0.168\ \frac{g}{L} NaHCO_3 \qquad \text{(Equation 3-3)}$$

The above calculation provides the amount of $NaHCO_3$ added to 1L of water to create a solution of 100 mg/L as $CaCO_3$ alkalinity. For the remaining solutions of 10, 30, 75, and 200 mg/L as $CaCO_3$, the previous calculation was repeated with the respective alkalinity concentrations. For the 100 mg/L $CaCO_3$ sample, 168 g of $NaHCO_3$ was weighed and added to a 1L storage container. 1L of deionized water was then added to the solution and was thoroughly mixed to create the 100 mg/L as $CaCO_3$ alkalinity solution. The same procedure was followed to create the 10, 30, 75, and 200 mg/L as $CaCO_3$ solutions. The pH of each solution was recorded.

3.2.5 Alkalinity Measurement

Alkalinity was measured with a volumetric titration. The pH meter was calibrated. A 50 mL burette was filled with 0.02N HCl and placed on a burette stand. 200 mL of the 100 mg/L as $CaCO_3$ alkalinity solution was measured using a 250 mL graduated cylinder and placed in a 400 mL beaker. A stir bar was added to the beaker and placed on a stir plate and the stir plate was turned on low. The pH probe was placed in the beaker and the initial pH was recorded. HCl titrant was slowly added to the solution and the pH was closely monitored. The solution was titrated to a pH of 4.5. The volume of titrant added to the sample was recorded and the alkalinity was calculated using the equation for alkalinity provided by Standard Methods as shown (APHA et al., 2017).

$$Alk = \frac{A \times N \times 50000}{mL\ NaHCO_3\ sample} \qquad \text{(Equation 3-4)}$$

In this equation, A is the volume of standard acid used and N is the normality of standard acid. The alkalinity titration was performed for each alkalinity sample and the alkalinities were calculated and recorded. The titrations were performed both at the beginning of the experiments as well as after the final samples were collected.

3.2.6 Lead

The aliquots were tested using an inductively coupled plasma-optical emission spectrometry (ICP-OES) instrument. Four lead solutions with hydrochloric acid for acidity were created in concentrations of 4, 20, 40, and 100 ppm to create the standard calibration curve. The aliquots, as well as the calibration curve solutions, were analyzed by a professional technician, and the results were provided to the MQP team.

4. Results and Discussion

Corrosion of lead coupons in waters with varying conditions were analyzed by measuring dissolved lead concentrations in the waters over time. Water quality conditions that were assessed included pH, alkalinity, and temperature. In this chapter, the stability of water quality parameters over time is discussed, followed by the impact of each parameter on dissolution of lead. The results are used to evaluate whether a short-term bench-scale test can be used to differentiate corrosion potential in waters.

4.1 pH

The impact of pH on corrosion was tested by creating samples with pH goals of 3.0, 6.5, 7.5, 8.0, and 12.0. Two samples were created at each pH condition, with one stored at 4°C and one stored at 20°C. To help keep pH stable, a phosphate buffer was used for the 6.5, 7.5, and 8.0 samples. The pH of the samples was measured at the beginning and end of the experiments and the resulting data can be seen in Table 3. For 8 out of 10 of the samples, the pH after four weeks remained within 1 pH unit of the starting conditions. Both samples with a pH goal of 3.0 changed more than 1 pH unit over the course of the 4-week period. These samples did not have any buffers to stabilize the pH. The sample with pH goal of 3.0 and a temperature of 20°C increased to a pH of 5.11, while the sample with pH goal of 3.0 and a temperature of 4°C increased to a pH of 4.60. Although these increases were more than 1 unit, the samples were still acidic and below the recommended pH range for water distribution systems. Therefore, the ten samples were suitable for assessing the impact of pH on corrosion as they properly represented low, middle, and high pH conditions during the testing period.

Table 3. pH Data on Samples with No Added Alkalinity

pH Goal	Temperature (°C)	Beginning pH	Ending pH
3.00	20	3.02	5.11
6.50	20	6.49	6.49
7.50	20	7.46	7.45
8.00	20	7.95	7.98
12.0	20	11.97	11.74
3.00	4.0	3.02	4.60
6.50	4.0	6.49	6.46
7.50	4.0	7.46	7.34
8.00	4.0	7.95	7.74
12.0	4.0	11.97	11.22

Ten additional samples were created to evaluate the impact of alkalinity on corrosion. These samples were created through the addition of sodium bicarbonate to distilled water with no pH adjustment. Sodium bicarbonate is a weak base, and therefore would be expected to result in a pH higher than 7. As shown in Table 4, the starting pH of these solutions ranged from 8.10 to 8.86 and therefore they were near the high end of the recommended range (6.5 – 8.5) for distribution systems. The pH values were also measured at the end of the 4-week period to see if the pH values were stable. Nine out of ten of the samples had pH values that remained within 1 pH unit of their starting value. One sample had a larger change: the sample with a starting alkalinity of 10 mg/L at 4°C had a starting pH of 8.86, which decreased to 7.83 after four weeks.

This ending pH value was still slightly basic and therefore was not of concern as all ten samples were representative of waters near the high end of the recommended pH range.

Table 4. pH Data on Samples with Added Alkalinity

Alkalinity (mg/L as $CaCO_3$)	Temperature (°C)	Beginning pH	Ending pH
10	20	8.86	9.14
30	20	8.10	9.01
75	20	8.19	8.99
100	20	8.23	8.54
200	20	8.29	8.59
10	4.0	8.86	7.83
30	4.0	8.10	7.96
75	4.0	8.19	7.97
100	4.0	8.23	7.89
200	4.0	8.29	8.08

4.2 Alkalinity

There were five different alkalinity concentrations tested, with the goals ranging from 10 to 200 mg/L as $CaCO_3$, and each alkalinity was tested at two different temperatures. Table 5 shows the alkalinity goal for each sample, and the measured alkalinity concentration after 4 weeks. Note that only the sample with a goal of 100 mg/L as $CaCO_3$ was tested at the start of the experiments. Through alkalinity titration, the measured alkalinity of this sample was 85.0 mg/L as $CaCO_3$. As all samples were created using a relative quantity of sodium bicarbonate compared to this sample, it was assumed that all samples had an actual starting concentration that was 15% less than the goal. These values are provided in Table 5 as the starting concentrations.

The alkalinity concentrations were measured for all 10 samples after 4 weeks. As shown in Table 5, a comparison of the ending concentrations and the actual starting concentrations demonstrated that the eight of the ten samples had alkalinity values that remained very consistent after 4-weeks (changes of a few percent increase or decrease). There were only two samples that had a substantial change in the measured alkalinity: 10 mg/L alkalinity sample at 4°C (94% increase in alkalinity) and the 10 mg/L alkalinity sample at 20°C (170% increase). It is possible that these samples had higher alkalinities at the start of the experiment than what was calculated. However, the ending alkalinities were still low (approximately 20 mg/L) and thus representative of low alkalinity waters. Therefore, the ten samples were suitable for assessing the impact of alkalinity on corrosion as they properly represented a range of alkalinity conditions during the testing period.

Table 5. Beginning and End Alkalinity Data

Alkalinity Goal (mg/L as $CaCO_3$)	Starting Alkalinity* (mg/L as $CaCO_3$)	Temperature (°C)	Ending Alkalinity (mg/L as $CaCO_3$)	Percent Change
10	8.5	20	16.5	+94%
30	25.5	20	25.5	no change
75	63.8	20	63.5	-0.5%
100	85.0	20	86.0	+1.2%
200	170	20	171	+0.6%
10	8.5	4	23.0	+170%
30	25.5	4	25.0	-2.0%
75	63.8	4	64.5	+1.1%%
100	85.0	4	86.5	+1.8%
200	170	4	176	+3.5%
* Value of 85 for samples with a goal of 100 was measured; all other values are calculated as a ratio of this measured value				

4.3 Lead

Corrosion of lead coupons in the water samples was assessed in two ways. First, dissolved lead concentrations were measured from aliquots collected from each water sample at time intervals of 1 day, 1 week, 3 weeks, and 4 weeks. Second, the weight of the coupons was measured at the start and end (4 weeks) of the experiments.

For the dissolved lead concentrations, lead was measured on an ICP-OES and the results were reported as ppb (µg/L) of lead. Out of the 80 samples, 17 had detectable levels of lead and 63 had lead concentrations below the detection limit (BDL) (see Tables 6-9). Of the 17 samples where lead was detected, 12 occurred in strong acid or strong base solutions. For example, for the strong base solution (pH goal = 12) at 20°C, the dissolved lead concentration was 4.4 µg/L after 1 day and 239 µg/L after 4 weeks. In comparison, the same strong base sample at 4°C had a lead concentration of BDL after 1 day and of 69.8 µg/L after 4 weeks.

The solutions with alkalinity resulted in five samples where lead was detected. The concentrations of lead that were detected in these samples were 45.0 µg/L, 6.95 µg/L, and 1.31 µg/L in the 30, 75, and 100 mg/L as $CaCO_3$ samples at 20°C, and 11.2 µg/L and 6.25 µg/L in the 10 and 75 mg/L as $CaCO_3$ samples at 4°C, respectively. All of these lead detections occurred in the third or fourth week, and no lead was detected in any of the samples with alkalinity after 1 day or 1 week.

Table 6. Lead Concentration in ppb With Varying pH at 20°C

pH Goal	1 Day	1 Week	3 Weeks	4 Weeks
3.0	5.55	95.0	118	111
6.5	BDL	BDL	BDL	BDL
7.5	BDL	BDL	BDL	BDL
8.0	BDL	BDL	BDL	BDL
12	4.38	BDL	239	239

Table 7. Lead Concentration in ppb With Varying pH at 4°C

pH Goal	1 Day	1 Week	3 Weeks	4 Weeks
3.0	BDL	32.9	148	133
6.5	BDL	BDL	BDL	BDL
7.5	BDL	BDL	BDL	BDL
8.0	BDL	BDL	BDL	BDL
12	BDL	BDL	96.8	69.8

Table 8. Lead Concentration in ppb with Varying Alkalinity at 20°C

Alkalinity Goal (mg/L $CaCO_3$)	1 Day	1 Week	3 Weeks	4 Weeks
10	BDL	BDL	BDL	BDL
30	BDL	BDL	BDL	45.0
75	BDL	BDL	BDL	6.95
100	BDL	BDL	BDL	1.31
200	BDL	BDL	BDL	BDL

Table 9. Lead Concentration in ppb With Varying Alkalinity at 4°C

Alkalinity Goal (mg/L $CaCO_3$)	1 Day	1 Week	3 Weeks	4 Weeks
10	BDL	BDL	11.2	BDL
30	BDL	BDL	BDL	BDL
75	BDL	BDL	BDL	6.25
100	BDL	BDL	BDL	BDL
200	BDL	BDL	BDL	BDL

In addition to measuring dissolved lead in the samples, the appearance of the water samples and the coupons was noted. The water samples were all clear when the experiments started. Over time, the appearance of the waters changed with regard to clarity. Figure 4 shows the sample with a pH goal of 3.0 stored at 20°C after 4 weeks of incubation. A white precipitate is apparent, making the water cloudy. Figure 5 shows a sample with an alkalinity goal of 10 mg/L as $CaCO_3$ that was stored at 20°C after 4 weeks of incubation. Again, a white precipitate can be seen in the solution; however, the precipitate is present in a much lower concentration than the sample with a pH goal of 3.0, so the water is clearer. Figure 6 shows the sample with pH goal of 7.5 stored at 20°C after 4 weeks of incubation. There is no precipitate present, so the water is clear. Considering all samples, the conditions in which precipitate formed were: pH goal of 3.0 at 20°C, alkalinity 10 mg/L at 20°C and 4°C, alkalinity at 30 mg/L at 20°C and 4°C, and alkalinity at 75 mg/L at 20°C. For all other conditions, the waters appeared clear after 4 weeks.

Figure 4. Appearance of sample with pH goal of 3.0 stored at 20°C after 4 Weeks

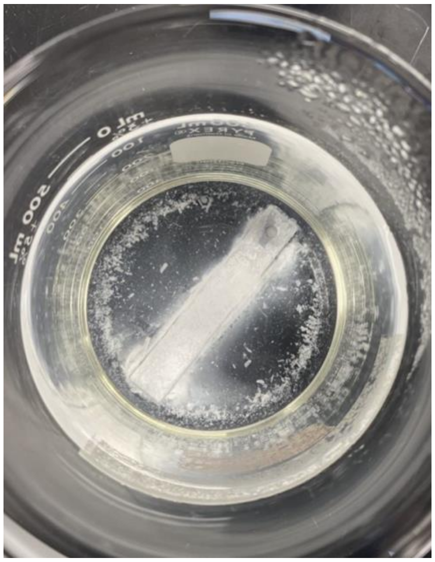

Figure 5. Appearance of sample with Alkalinity goal of 10 mg/L as $CaCO_3$ stored at 20°C after 4 Weeks

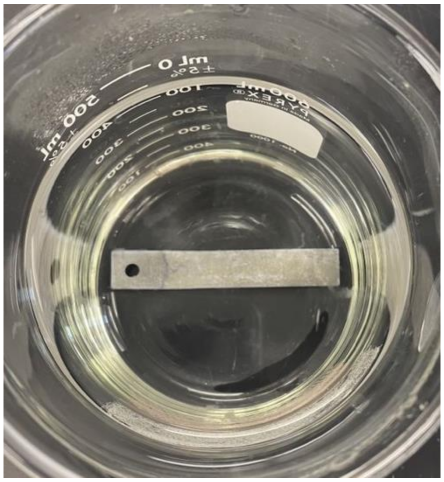

Figure 6. Appearance of sample with pH goal of 7.5 stored at stored at 20°C after 4 Weeks

The appearance of the coupons was observed at the conclusion of the four weeks of testing. The coupons were removed from the water samples, air dried, and then visually observed as well as weighed. Some coupons experienced little to no change in appearance; some had a slight white tint; and some showed drastic changes in color (e.g., rust-colored, yellow, or white). All the coupons are shown in Figure 7.

Figure 7. All Coupons (Left to Right, Top to Bottom: Alkalinity Goal of 10, 30, 75, 100, and 200 at 20°C, Alkalinity Goal of 10, 30, 75, 100, and 200 at 4°C, pH Goal of 3.0, 6.5, 7.5, 8.0, and 12.0 at 20°C, pH Goal of 3.0, 6.5, 7.5, 8.0, and 12.0 at 4°C) After 4 Weeks of Incubation

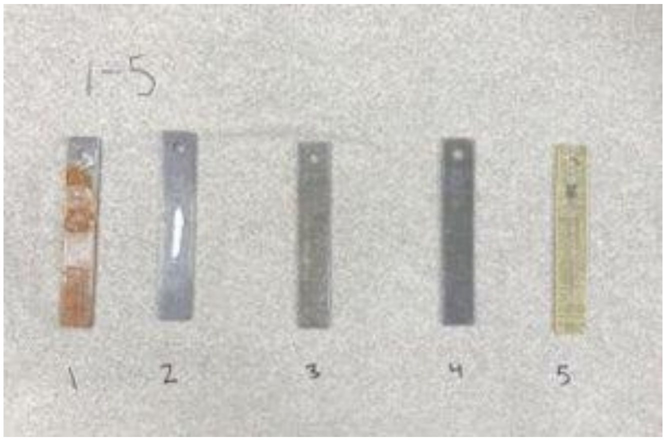

Figure 8. Coupons with Varying pH (Left to Right, pH Goal of 3.0, 6.5, 7.5, 8.0, and 12.0) at 20°C After 4 Weeks of Incubation

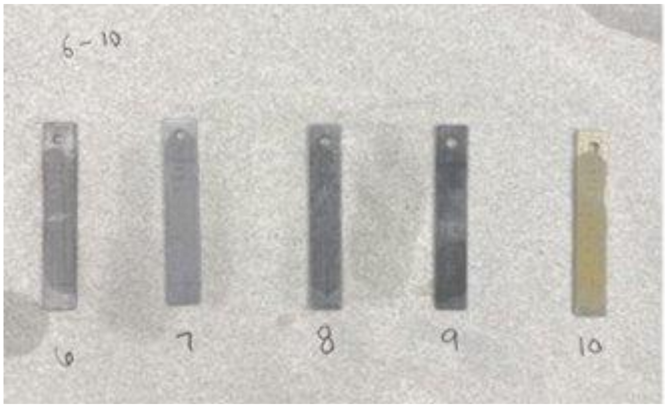

Figure 9. Coupons with Varying pH (Left to Right, pH Goal of 3.0, 6.5, 7.5, 8.0, and 12.0) at 4°C After 4 Weeks of Incubation

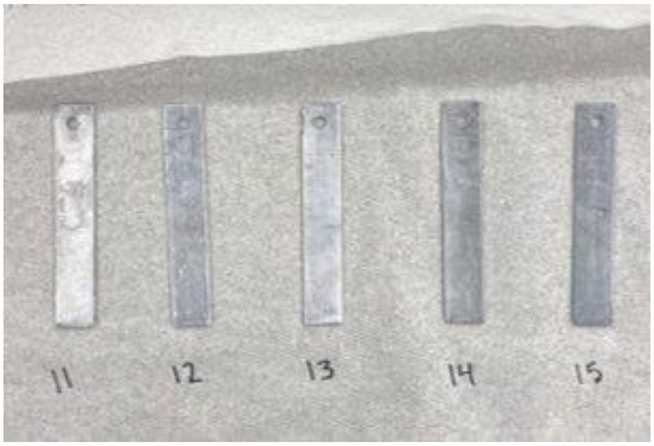

Figure 10. Coupons with Varying Alkalinity (Left to Right, Alkalinity Goal of 10, 30, 75, 100, and 200) at 20°C After 4 Weeks of Incubation

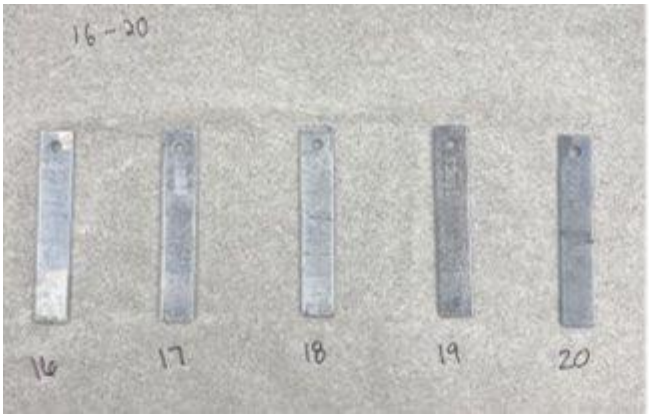

Figure 11. Coupons with Varying Alkalinity (Left to Right, Alkalinity Goal of 10, 30, 75, 100, and 200) at 4°C After 4 Weeks of Incubation

As seen in Figures 8-11 above, coupons 1-10 (with varying pH and no alkalinity) showed a greater diversity of appearance after 4 weeks than coupons 11-20 (with varying alkalinity at slightly alkaline pH). As stated previously, samples 1 (pH goal 3.0, 20°C), 5 (pH goal 12.0, 20°C), 6 (pH goal 3.0, 4°C) and 10 (pH goal 12.0, 4°C) had dissolved lead detected using an ICP-OES instrument. Coupons 1, 5 and 10 visually appear to have changed the most, and thus the visual changes are consistent with the lead results. Figures 10 and 11 show the coupons that were placed in waters with different alkalinity levels. These coupons did not change as much in appearance, with most of the changes being the coupons turning white and/or getting a white cloudy look. This is consistent with precipitation of carbonates, which can form white solids. These changes also align with the results from the ICP-OES, in which dissolution of lead was not observed until the third or fourth week of incubation and concentrations of lead in solutions with alkalinity were generally lower than in solutions without alkalinity. Additional photos of the lead coupons after the 4-week period can be found in Appendix 7.3.

In addition to the ICP-OES results, each coupon was weighed after being placed on a paper towel to dry. The results are shown in Table 10, where 18 of the 20 coupons went down in weight over the four weeks. The two coupons that increased in weight (coupons 2 and 3) had very minor increases of thousandths of a percent. The coupons with the largest dissolved lead concentrations per ICP-OES results had larger weight changes, especially coupons 1, 5, 6, and 10 which represented the extreme pH conditions. These results affirm one another, since the coupons were corroding in the water and having lead dissolved, making the coupons weigh less, and making the waters have detectable dissolved lead concentrations.

Table 10. Starting and Ending Weights of the Lead Coupons

Coupon Number-Jar	Test Conditions (pH or alkalinity goal and temperature)	Starting weight (mg)	End weight (mg)	Change in weight (%)
5377-1	3.0 pH, 20°C	16149.5	16053.8	-0.593%
5378-2	6.5 pH, 20°C	16282.0	16282.4	+0.002%
5379-3	7.5 pH, 20°C	16278.5	16278.0	+0.003%
5380-4	8.0pH, 20°C	16177.0	16175.0	-0.012%
5381-5	12.0 pH, 20°C	16312.0	16189.5	-0.761%
5382-6	3.0 pH, 4°C	16197.0	16127.9	-0.427%
5383-7	6.5 pH, 4°C	16257.7	16256.8	-0.006%
5384-8	7.5 pH, 4°C	16189.0	16185.2	-0.024%
5385-9	8.0 pH, 4°C	16262.0	16258.6	-0.021%
5386-10	12.0 pH, 4°C	16311.5	16267.1	-0.273%
5387-11	10 mg/L $CaCO_3$, 20°C	16359.0	16329.3	-0.182%
5388-12	30 mg/L $CaCO_3$, 20°C	16117.0	16112.7	-0.027%
5389-13	75 mg/L $CaCO_3$, 20°C	16333.5	16327.3	-0.038%
5390-14	100 mg/L $CaCO_3$, 20°C	16197.5	16193.9	-0.023%
5391-15	200 mg/L $CaCO_3$, 20°C	16173.5	16169.9	-0.023%
5392-16	10 mg/L $CaCO_3$, 4°C	16242.5	16221.0	-0.133%
5393-17	30 mg/L $CaCO_3$, 4°C	16294.0	16281.7	-0.076%
5394-18	75 mg/L $CaCO_3$, 4°C	16292.0	16286.7	-0.033%
5395-19	100 mg/L $CaCO_3$, 4°C	16280.5	16275.5	-0.031%
5396-20	200 mg/L $CaCO_3$, 4°C	16123.0	16116.4	-0.041%

4.3.1 Temperature Impact on Lead Corrosion

Corrosion of metals is known to increase with increasing temperature. In general, for every 10°C increase in temperature, the rate of corrosion doubles. In the bench-scale tests, dissolved lead was detected in samples earlier, more frequently, and at higher concentrations when the samples were incubated at 20°C compared to samples incubated at 4°C. However, this generalization did not hold for all sample conditions.

Figures 12a through 12d show dissolved lead concentrations for samples with varying pH and varying temperature, where each graph provides data for one of the sampling times. Note that Figure 12a has a different y-axis scale than Figures 12b-d. In each graph, detectable levels of lead are shown in blue for samples at 20°C, and in orange for samples at 4°C. In Figure 12a after 1 day of incubation, the extreme pH samples (3.0 and 12.0) have detectable lead but only for the higher temperature samples. After 1 week (Figure 12b), the acidic sample (pH 3.0) has detectable lead at both temperatures, but the concentration at the higher temperature (95.0 µg/L) is almost three times the concentration at the lower temperature (32.9 µg/L). After 3 weeks (Figure 12c) and 4 weeks (Figure 12d), the lead concentrations in the pH 3.0 samples are similar at both temperatures, but for the pH 12.0 samples, the lead concentrations are much higher at 20°C (239 µg/L) than at 4°C (69.8 µg/L). For samples with varying alkalinity (data shown previously in Tables 8 and 9), samples after 4 weeks of incubation had higher lead levels at 20°C compared with 4°C, noting that these samples had very low levels of dissolved lead overall.

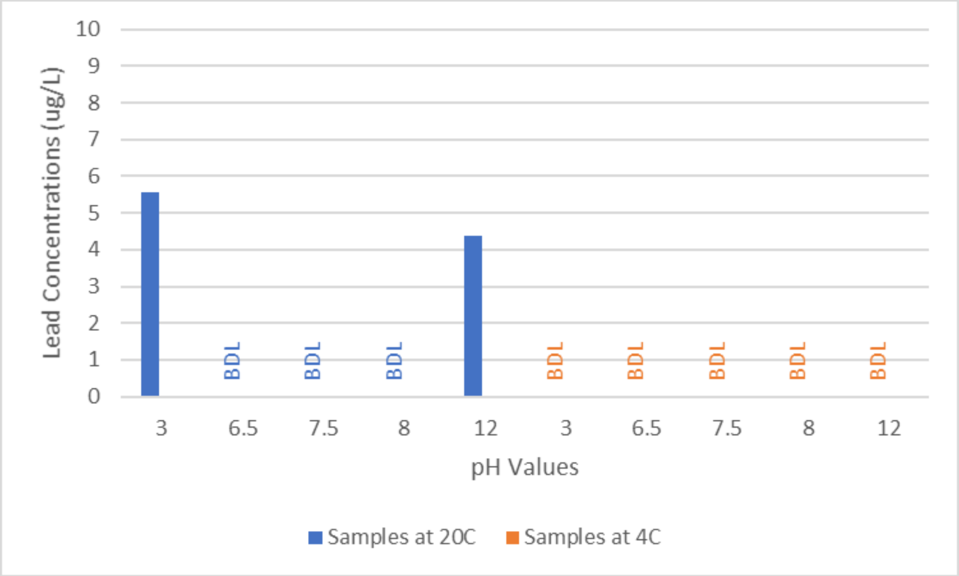

Figure 12a. Effect of Temperature on Lead Concentrations for Samples with Varying pH after 1 day (note y-axis scale maximum is 10 compared to Figures 12b, c, and d maximum of 300)

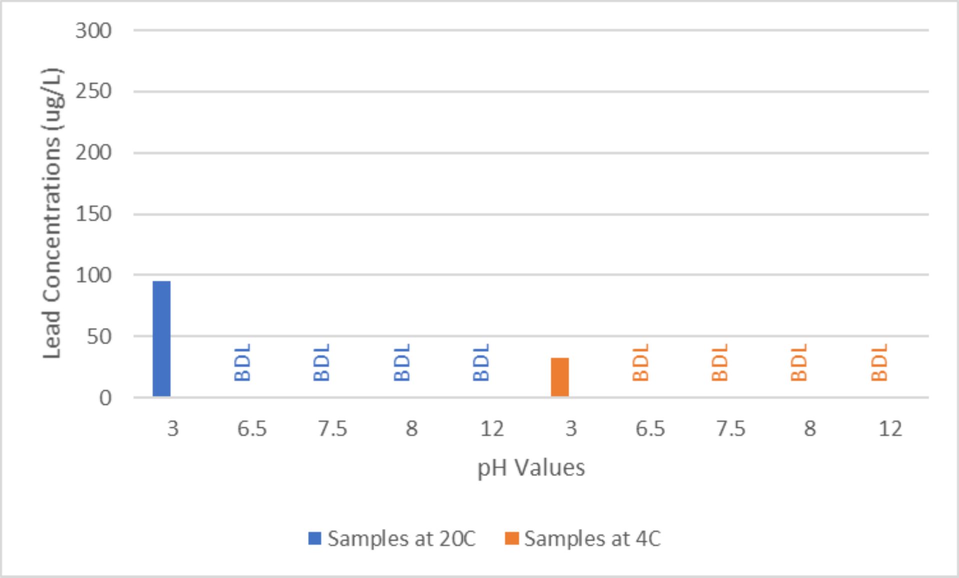

Figure 13b. Effect of Temperature on Lead Concentrations for Samples with Varying pH after 1 week

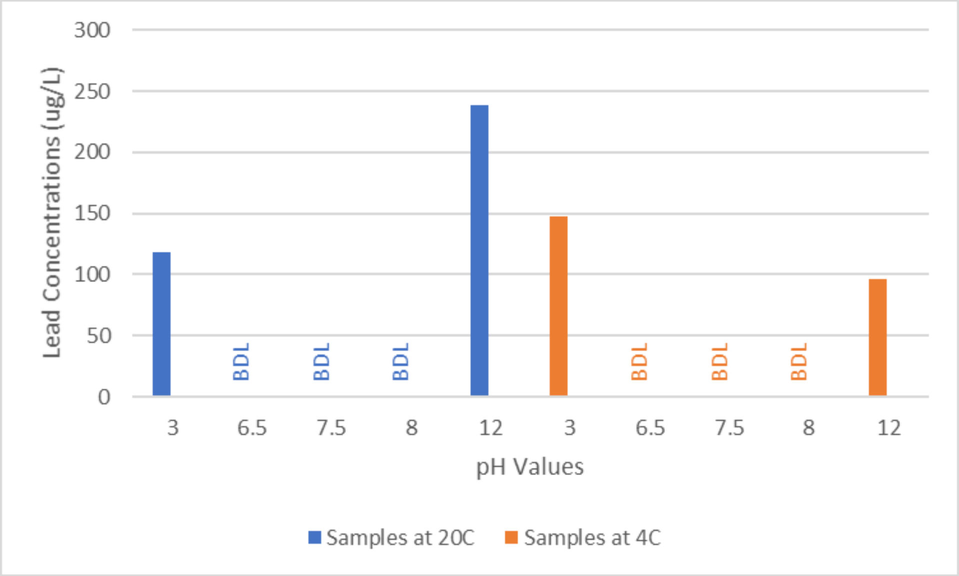

Figure 14c. Effect of Temperature on Lead Concentrations for Samples with Varying pH after 3 weeks

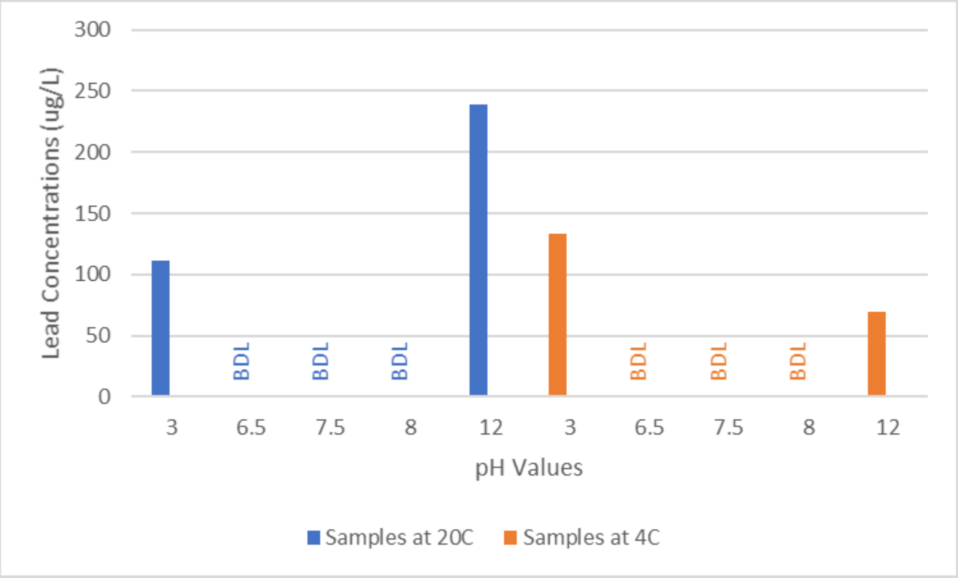

Figure 15d. Effect of Temperature on Lead Concentrations for Samples with Varying pH after 4 weeks

4.3.2 pH Impact on Lead Corrosion

The pH of the solutions had a significant impact on the detection of lead, as well as the concentration of dissolved lead. There was no dissolved lead detected in any samples for which the pH goal was 6.5, 7.5, or 8.0, regardless of the incubation temperature and incubation time. In contrast, the strong acid (pH goal = 3.0) and strong base solutions (pH goal = 12.0) had dissolved lead as early as day one. As shown in Figure 13, samples incubated at 20°C had lead concentrations of 4-6 µg/L after 1 day, and lead concentrations in the hundreds of µg/L after 3 or 4 weeks. Samples incubated at 4°C showed similar patterns of increasing dissolved lead over time, although the amount of corrosion was overall less than in the higher temperature samples. These results indicate that pH conditions that fall within the recommended pH values for drinking water distribution systems did not result in measurable corrosion of lead coupons over a 4-week test, while extreme pH conditions did result in detectable corrosion of the coupons with expected patterns of increasing corrosion over time. Therefore, if a utility wants to compare pH conditions within the range of 6.5 to 8.0, a short-term test may not provide useful data.

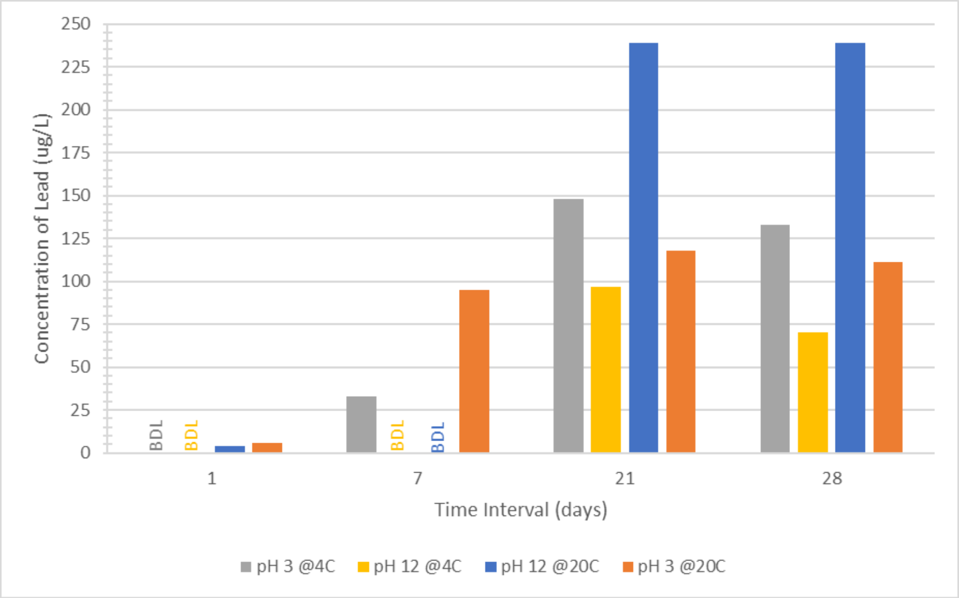

Figure 16. Lead Concentration Over Time with Regards to pH and Temperature

4.3.3 Alkalinity Impact on Corrosion

Alkalinity values from 10 to 200 mg/L as $CaCO_3$ were compared for their effect on the dissolution of lead. All samples incubated for 1 day or 1 week, regardless of temperature, had undetectable levels of lead. There were some detectable levels of lead after 3 or 4 weeks of incubation time; however, a pattern was not obvious. Figure 14 shows the results for lead concentrations after 3 and 4 weeks of incubation time. As shown, one sample (30 mg/L as $CaCO_3$, 20°C, 4 weeks) had a lead concentration of approximately 45 µg/L, and several samples had lead concentrations less than or near 10 µg/L. Detectable lead was generally associated with lower alkalinity. However, the sample with 30 mg/L as $CaCO_3$ alkalinity had the highest lead concentration in week 4, and the sample with 10 mg/L as $CaCO_3$ alkalinity had detectable lead in week 3 but not in week 4. Overall, the results do not indicate that a short-term (4-week) test can adequately differentiate corrosion potential in samples with moderate levels of alkalinity that would be expected in a drinking water distribution system.

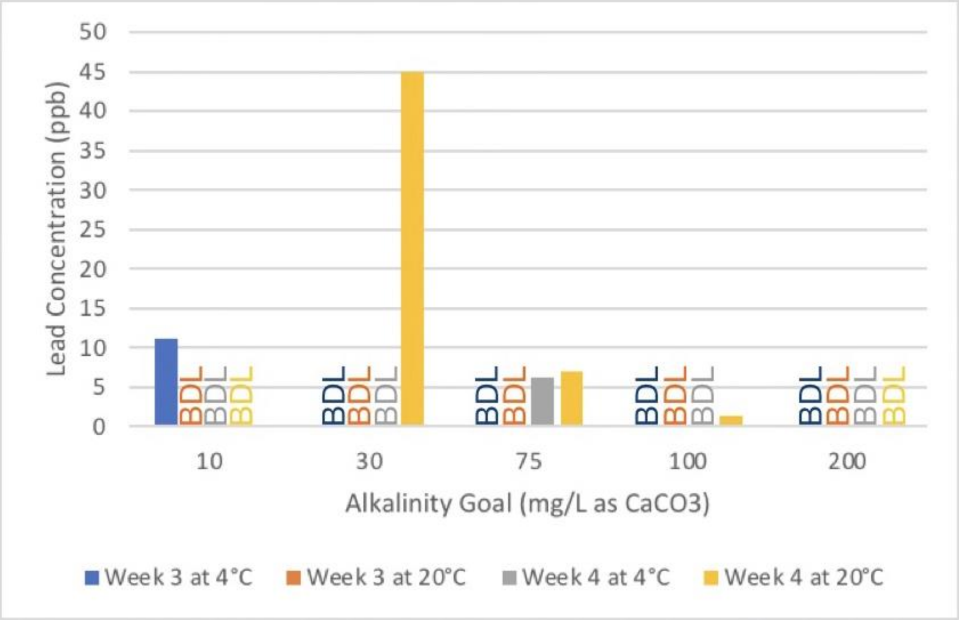

Figure 17. Alkalinity Impact on Lead Concentration

4.4 Analysis of Lead Concentrations

The samples that had the greatest concentration of lead were the strong acid and base samples. The strong acid and base samples, regardless of temperature, resulted in high concentrations of lead. The sample with a pH of 12 at 20°C resulted in the greatest concentration of lead (239 µg/L), while the sample with a pH of 12 at 4°C still had a high lead concentration (69.8 µg/L). For the samples at pH 3.0, both high and low temperatures resulted in lead concentrations over 100 µg/L after 4 weeks. For these acidic samples, pH had fluctuated over the course of the experiment such that the pH in the colder samples was 0.5 units below the pH in the warmer samples, and this may be the reason why the colder sample had a higher lead concentration (133 versus 111 µg/L). While some of the alkalinity samples had lead detection, the concentrations were generally low and occurred primarily in the day 28 samples, with the highest lead concentration in one of the samples with low alkalinity (30 mg/L as $CaCO_3$).

There is potential for a short-term test to differentiate corrosion potential. This test was most effective with waters that have an extreme pH. However, for waters with moderate pH conditions and moderate alkalinity, results were less conclusive due to low or non-detectable lead concentrations.

5. Conclusion and Recommendations

Experimental results demonstrated that a short-term, bench-scale test using lead coupons immersed in water samples is able to differentiate lead corrosion potential in waters with extreme pH conditions compared to waters with near neutral pH conditions. In the extreme pH conditions, lead concentrations generally increased over time and with temperature, while lead was not detected in samples with near neutral pH conditions. The conditions likely to be seen in a water distribution system (near neutral pH and moderate alkalinity) showed no or very little corrosion, which is desirable for a full-scale system but also did not allow for quantitative comparison of such conditions at the bench-scale.

The instrument used for measuring lead concentrations in this project was an ICP-OES due to unforeseen circumstances with the availability of the ICPMS. The use of the ICPMS would likely have been able to detect lead concentrations in smaller amounts and therefore may have been able to detect lead in the more moderate waters where the lead did not corrode as much as the extreme conditions. The test that the team conducted was done using stagnant water when in an actual distribution system, the water would likely be moving. When the water is moving, it causes more disruption against the pipe, which could result in more corrosion of the lead pipe. Had the experiment been conducted using a constant mixer, it could have increased the amount of corrosion in each sample and therefore the test may have been able to detect lead present in more moderate samples.

The test might also benefit from an extension of time. It is possible that an additional week or two could result in more corrosion that could be recognized by the test. However, the purpose of the test was to be more timely and less expensive, and by extending the length of the test, it is possible that it will no longer achieve this goal. Because of this, it is recommended that other factors, such as instrumentation used to measure lead concentrations and constant movement of the water, be explored first before extending the length of the test.

Since there are multiple unknowns with this short-term test, the pipe-loop and coupon test remain more reliable in determining lead corrosion until the short-term lab test can be improved. The tests might be more expensive and time intensive, but they are better equipped to differentiate corrosion potential in waters that are representative of distribution system conditions. The consumption of lead in any amount is toxic to the human body, especially infants, children under 6, and pregnant women, so an accurate test is of the utmost importance. The protection of public health is worth the extra cost of time and money until a more robust short-term test can be created.

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7. Appendices

7.1 Lead concentration over time with varying pH

Jar	pH Goal	Temperature (°C)	Time of Extraction	Lead Concentration (µg/L)
1 – 1 Day	3.0	20	1 Day	5.55
1 – 1 Week	3.0	20	1 Week	95.0
1 – 3 Weeks	3.0	20	3 Weeks	118
1 – 4 Weeks	3.0	20	4 Weeks	111
2 – 1 Day	3.0	20	1 Day	Non-detect
2 – 1 Week	3.0	20	1 Week	Non-detect
2 – 3 Weeks	3.0	20	3 Weeks	Non-detect
2 – 4 Weeks	3.0	20	4 Weeks	Non-detect
3 – 1 Day	6.5	20	1 Day	Non-detect
3 – 1 Week	6.5	20	1 Week	Non-detect
3 – 3 Weeks	6.5	20	3 Weeks	Non-detect
3 – 4 Weeks	6.5	20	4 Weeks	Non-detect
4 – 1 Day	6.5	20	1 Day	Non-detect
4 – 1 Week	6.5	20	1 Week	Non-detect
4 – 3 Weeks	6.5	20	3 Weeks	Non-detect
4 – 4 Weeks	6.5	20	4 Weeks	Non-detect
5 – 1 Day	7.5	20	1 Day	Non-detect
5 – 1 Week	7.5	20	1 Week	Non-detect
5 – 3 Weeks	7.5	20	3 Weeks	Non-detect
5 – 4 Weeks	7.5	20	4 Weeks	Non-detect
6 – 1 Day	7.5	4	1 Day	Non-detect
6 – 1 Week	7.5	4	1 Week	32.9
6 – 3 Weeks	7.5	4	3 Weeks	148
6 – 4 Weeks	7.5	4	4 Weeks	133
7 – 1 Day	8.0	4	1 Day	Non-detect
7 – 1 Week	8.0	4	1 Week	Non-detect
7 – 3 Weeks	8.0	4	3 Weeks	Non-detect
7 – 4 Weeks	8.0	4	4 Weeks	Non-detect
8 – 1 Day	8.0	4	1 Day	Non-detect
8 – 1 Week	8.0	4	1 Week	Non-detect
8 – 3 Weeks	8.0	4	3 Weeks	Non-detect
8 – 4 Weeks	8.0	4	4 Weeks	Non-detect
9 – 1 Day	12	4	1 Day	4.38
9 – 1 Week	12	4	1 Week	Non-detect
9 – 3 Weeks	12	4	3 Weeks	239
9 – 4 Weeks	12	4	4 Weeks	239
10 – 1 Day	12	4	1 Day	Non-detect
10 – 1 Week	12	4	1 Week	Non-detect

10 – 3 Weeks	12	4	3 Weeks	96.8
10 – 4 Weeks	12	4	4 Weeks	69.8

7.2 Lead concentration over time with varying alkalinity

Jar	Alkalinity Goal (mg/L as $CaCO_3$)	Temperature (°C)	Time of Extraction	Lead Concentration (µg/L)
11 – 1 Day	10	20	1 Day	Non-detect
11 – 1 Week	10	20	1 Week	Non-detect
11 – 3 Weeks	10	20	3 Weeks	Non-detect
11 – 4 Weeks	10	20	4 Weeks	Non-detect
12 – 1 Day	30	20	1 Day	Non-detect
12 – 1 Week	30	20	1 Week	Non-detect
12 – 3 Weeks	30	20	3 Weeks	Non-detect
12 – 4 Weeks	30	20	4 Weeks	45.0
13 – 1 Day	75	20	1 Day	Non-detect
13 – 1 Week	75	20	1 Week	Non-detect
13 – 3 Weeks	75	20	3 Weeks	Non-detect
13 – 4 Weeks	75	20	4 Weeks	6.95
14 – 1 Day	100	20	1 Day	Non-detect
14 – 1 Week	100	20	1 Week	Non-detect
14 – 3 Weeks	100	20	3 Weeks	Non-detect
14 – 4 Weeks	100	20	4 Weeks	1.31
15 – 1 Day	200	20	1 Day	Non-detect
15 – 1 Week	200	20	1 Week	Non-detect
15 – 3 Weeks	200	20	3 Weeks	Non-detect
15 – 4 Weeks	200	20	4 Weeks	Non-detect
16 – 1 Day	10	4	1 Day	Non-detect
16 – 1 Week	10	4	1 Week	Non-detect
16 – 3 Weeks	10	4	3 Weeks	11.2
16 – 4 Weeks	10	4	4 Weeks	Non-detect
17 – 1 Day	30	4	1 Day	Non-detect
17 – 1 Week	30	4	1 Week	Non-detect
17 – 3 Weeks	30	4	3 Weeks	Non-detect
17 – 4 Weeks	30	4	4 Weeks	Non-detect
18 – 1 Day	75	4	1 Day	Non-detect
18 – 1 Week	75	4	1 Week	Non-detect
18 – 3 Weeks	75	4	3 Weeks	Non-detect
18 – 4 Weeks	75	4	4 Weeks	6.25
19 – 1 Day	100	4	1 Day	Non-detect
19 – 1 Week	100	4	1 Week	Non-detect
19 – 3 Weeks	100	4	3 Weeks	Non-detect

19 – 4 Weeks	100	4	4 Weeks	Non-detect
20 – 1 Day	200	4	1 Day	Non-detect
20 – 1 Week	200	4	1 Week	Non-detect
20 – 3 Weeks	200	4	3 Weeks	Non-detect
20 – 4 Weeks	200	4	4 Weeks	Non-detect

7.3 Lead Coupon Photos

Figure 18. Coupon 5377 at 4 Weeks with pH 3 and Temperature 20°C

Figure 19. Coupon 5378 at 4 Weeks with pH 6.5 and Temperature 20°C

Figure 20. Coupon 5379 at 4 Weeks with pH 7.5 and Temperature 20°C

Figure 21. Coupon 5380 at 4 Weeks with pH 8 and Temperature 20°C

Figure 22. Coupon 5381 at 4 Weeks with pH 12 and Temperature 20°C


Figure 23. Coupon 5382 at 4 Weeks with pH 3 and Temperature 4°C

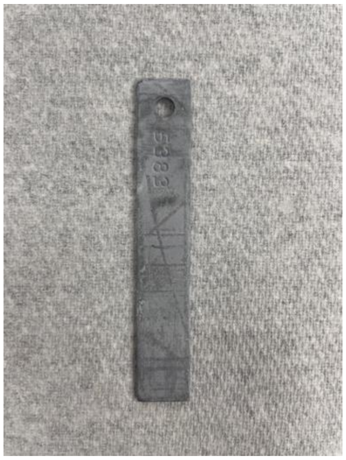
Figure 24. Coupon 5383 at 4 Weeks with pH 6.5 and Temperature 4°C


Figure 25. Coupon 5384 at 4 Weeks with pH 7.5 and Temperature 4°C

Figure 26. Coupon 5385 at 4 Weeks with pH 8 and Temperature 4°C

Figure 27. Coupon 5386 at 4 Weeks with pH 12 and Temperature 4°C

Figure 28. Coupon 5387 at 4 Weeks with Alkalinity 10 and Temperature 20°C

Figure 29. Coupon 5388 at 4 Weeks with Alkalinity 30 and Temperature 20°C

Figure 30. Coupon 5389 at 4 Weeks with Alkalinity 75 and Temperature 20°C

Figure 31. Coupon 5390 at 4 Weeks with Alkalinity 100 and Temperature 20°C

Figure 32. Coupon 5391 at 4 Weeks with Alkalinity 200 and Temperature 20°C

Figure 33. Coupon 5392 at 4 Weeks with Alkalinity 10 and Temperature 4°C

Figure 34. Coupon 5393 at 4 Weeks with Alkalinity 30 and Temperature 4°C

Figure 35. Coupon 5394 at 4 Weeks with Alkalinity 75 and Temperature 4°C

Figure 36. Coupon 5395 at 4 Weeks with Alkalinity 100 and Temperature 4°C

Figure 37. Coupon 5396 at 4 Weeks with Alkalinity 200 and Temperature 4°C