Impact of Chloride: Sulfate Mass Ratio (CSMR) Changes on Lead Leaching in Potable Water [Project #4088]

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OBJECTIVES:
The Water Research Foundation funded this project to determine the potential effects of coagulant changeover on lead release from plumbing components, with emphasis on changeover from aluminum sulfate to ferric chloride, ferric sulfate, polyaluminum chloride, and various polymer coagulants. Further, project goals were to investigate the finished water chemistry resulting from coagulant changeover, how distribution system scales are affected and how this affects lead release, and the magnitude of lead release after different stagnation times.

BACKGROUND:
The leaching of lead to potable water can sometimes be severely impacted by seemingly innocuous changes in water treatment including changes from one coagulant type to another. Case studies have documented a few prior instances in which coagulant changes produced a higher chloride-sulfate mass ratio (CSMR) in the finished water that was linked to the onset of lead contamination problems. However, no mechanistic studies had been conducted to unambiguously demonstrate the precise sequence of events that can cause higher a CSMR to increase lead contamination.

APPROACH:
The work was performed in three phases. In Phase 1, the fundamental chemistry of galvanic corrosion attack on lead-copper joints was evaluated, and experiments examined impacts of high CSMR on the integrity of soldered joints. Utility case studies were evaluated in the second phase of work to examine effects of CSMR on galvanic corrosion in a number of potable waters. Specifically, questions regarding the effects of coagulant changeover, desalination, and anion exchange treatment on lead solder and leaded brass corrosion were evaluated. The roles of alkalinity, pH, and corrosion inhibitors to potentially mitigate corrosion in high CSMR waters were also examined. Finally, in a third phase of work, re-circulating loops were used to evaluate the impacts of chloride, sulfate, and flow rate on corrosion of lead plumbing materials.

RESULTS/CONCLUSIONS:
• A simple bench-scale test protocol was developed that provides insights to changes in lead leaching to water from galvanic connections between solder/lead pipe and copper. This simple dump and fill protocol was successfully used to rapidly screen for significant changes in lead leaching that resulted from various water treatment changes. In most cases, short-term tendencies as measured by the test were in qualitative agreement with practical utility experiences and longer-term test results.
• pH drops at the surface of lead solder or lead pipe, if there is a galvanic connection to copper pipe. pHs as low as 3.3 were measured at the surface of the lead bearing material even when the bulk water pH was 8.0 or higher. (Chapter 1)
• At pH 3, pH 4, and pH 5, the concentration of soluble lead decreased with the addition of high levels of sulfate. Changes in pH in the pH 3–5 range have little effect on the solubility of lead sulfate. The migration of sulfate to lead anode surfaces can therefore serve to limit lead leaching. (Chapter 1)
• At pH 3, 4, and 5, the uncomplexed or free lead concentration decreases with the addition of chloride, consistent with formation of a PbCl+ lead chloride complex. The migration of chloride to lead anode surfaces, therefore, can exacerbate problems with lead leaching. (Chapter 1)
• The net effect of chloride and sulfate migration to lead anode surfaces depends on their relative concentration. Higher chloride tends to increase lead solubility whereas higher sulfate tends to decrease lead solubility; hence, the usefulness of the CSMR in explaining trends in lead contamination when galvanic corrosion of lead is significant. (Chapter 1)

• The corrosion rate of and the release of lead and/or tin from solder alloys was greater in high CSMR water. The pH at the solder surfaces was measured to be as low as pH 3.0. (Chapter 2)

• Simulated joints with 97/3 Sn/Cu solder had the greatest reduction of joint strength after one year of exposure to high CSMR water. (Chapter 2)

• The 95/5 Sn/Sb had the most desirable characteristics of the solders evaluated in this project. The solder had the lowest reduction in joint strength after one year of exposure to high CSMR water, did not release harmful levels of antimony, and had less tin corrosion compared to other solder alloys. (Chapter 2)

• For the utilities evaluated in this project, leaded brass leached relatively low levels of lead to the water, even in situations with high CSMR. In contrast, corrosion of lead solder in simulated copper joints contributed to very high amounts of lead in test waters. Thus, while leaded brass is impacted somewhat by CSMR, the issues associated with lead solder can occasionally achieve hazardous waste levels (>5,000 ppb) of lead in water under worst-case scenarios. As a result, lead solder and lead pipe galvanically connected to copper are the primary concern when effects of higher CSMR are considered.

• Generally, increasing the chloride-to-sulfate mass ratio (CSMR) of the water results in higher lead levels in water when copper:lead solder or copper:lead pipe galvanic couples are present. There could be higher chloride and lower sulfate in the water due to a range of scenarios:
  - Road salt entering the water supply from runoff, especially into open reservoirs (Chapter 1)
  - Coagulant type (chloride-based vs. sulfate-based) (Chapters 4, 5, 6, and 7)
  - Desalination (Chapter 8)
  - Chloride-based anion exchange treatment (Chapter 6 and 9)
  - Brine leak from hypochlorite generation system (Chapter 10)

• A combination of low pH and high CSMR at the solder surface drive the long-term galvanic corrosion of the lead solder, as demonstrated by local measurements of very high chloride, sulfate, and lead near the lead solder anode. There is no evidence that the lead solder can passivate in such circumstances.

• It is hypothetically possible that in systems with high CSMR, much of the solder that was exposed to water has been released over decades of corrosion. A substantial fraction of the simulated solder from worst-case joints leached to the water over a period of months to years. Hence, such systems might not be adversely impacted by higher CSMR water, relative to systems that passivated rapidly in a low CSMR water when first installed and abruptly subjected to high CSMR water. This might also explain compliance with 90th percentile lead levels in systems that have historically had high CSMR water.

• Increasing alkalinity of the water can be effective in mitigating the low pH at the lead solder surface and eventually causing decreased lead levels. However, in some circumstances where conductivity of the water is very low, increasing the alkalinity (and conductivity) can make lead leaching much worse. Dosing orthophosphate was more effective than polyphosphate in reducing lead release. However, in rare circumstances, dosing orthophosphate could make lead leaching worse. Hence, the effects of phosphate and alkalinity need to be determined on a site-specific basis. With additional experience, it might be discovered that the adverse effects of higher alkalinity and higher phosphate are limited to certain waters, such as those with extremely low conductivity. (Chapters 6 and 10)

• The effect of CSMR was confirmed in flowing conditions that are typical in home systems. Higher CSMR water resulted in higher lead leaching from lead pipe, bronze pipe, and solder galvanically connected to copper. (Chapter 11)
• The galvanic connection of copper to the lead materials evaluated in the study significantly increased lead leaching when compared to the situation when there was no electrical connection to copper pipe. In some waters, however, galvanic connections had little effect on lead leaching. (Chapter 11)

APPLICATIONS/RECOMMENDATIONS:
A key finding was that problems that occur in coagulant changeovers could usually be mitigated by controlling the type of coagulant and keeping CSMR below about 0.5. However, this is not always an option when CSMR was increased via arsenic treatment via anion exchange or desalination. For these case studies, adding orthophosphate when the CSMR was high did not reduce lead leaching or the extent of the problem. Therefore, more work is needed to determine what treatment(s) could counter the adverse consequences of higher CSMR.

This work also focused on making comparisons among treatment methods in terms of leaching from lead solder under worst-case conditions of long stagnation times, and these changes were dramatic. However, it is not completely certain how the lead levels measured in these “worst case” scenarios would translate to 90th percentile lead in practice. A better understanding would require consideration of many factors including plumbing materials in the distribution system, variability in plumbing jobs, and other factors. That is, in some situations, even a 20% increase in lead leaching from solder or lead pipe connected to copper due to higher CSMR might translate to a relatively modest increase (+/- 0.1 to 1 ppb) in 90th percentile lead. In these cases, even though galvanic corrosion of lead bearing materials is worsened, the public health concern might be of relatively low significance.

A roadmap to the report is provided to assist utilities, regulators, and scientists in determining which portions of this work would be of greatest interest in their particular situation (see the report for Figure ES.2). Utilities with significant to serious lead concerns from CSMR, as determined from Figure ES.1 in the report, are strongly encouraged to consider the relevant portions of this work.

RESEARCH PARTNER:
U.S. Environmental Protection Agency

PARTICIPANTS:
Twelve utilities from the United States and Canada participated in this project.