# A Systematic Study on the Control of Lead in a New Building

Darren A Lytle, Environmental Engineer, Michael R. Schock, Research Chemist, and Thomas J. Sorg, Branch Chief

U.S. Environmental Protection Agency RREL, DWRD, 1&PCB Cincinnati, Ohio 45268

#### Background

In early 1991, a federal agency requested the assistance of the U.S. Environmental Protection Agency's (EPA's) Drinking Water Research Division (DWRD) to solve a problem of high lead in the drinking water of a research facility built to house approximately 1000 employees. Drinking water samples taken from various sites in their newly constructed facility contained extremely high levels of lead. The facility, whose construction was completed in 1986, had been unoccupied, with the exception of a few building maintenance employees, due to a series of structural defects and repairs.

About six months before the building was scheduled to be opened, the water in the building was tested for a range of regulated inorganic contaminants. Samples where taken from a variety of locations including water coolers, bathroom faucets, laboratory faucets, and sample bottle washer units. Results indicated that the water exceeded the lead maximum contaminant level (MCL) which at that time was 0.05 mg/L. The lead test results prompted a number of follow-up studies to establish the degree of lead contamination in the building's drinking water. Between December, 1990 and February, 1991, two more sampling studies were conducted. A wide variety of locations throughout the building were sampled and the samples were split between two laboratories. Test results indicated extremely high lead levels in many locations (as high as 1.4 mg/L). However inconsistencies among lead levels seen in samples taken from the same site on multiple sampling periods and split sample results from multiple laboratories raised questions regarding the testing and sampling protocols.

Because of high lead levels and the inconsistency of the sampling results, the EPA's Drinking Water Research Division was requested to visit the facility and re-sample the facility. DWRD sampled 19 of the previously tested locations using a slightly different sampling protocol than was used before. The new protocol consisted of taking 250 mL samples following a 10 minute flushing period and after a 24 hour standing time. This procedure was based on EPA's protocol for testing lead in drinking water in schools and buildings.<sup>1,2</sup> The protocol sets 20  $\mu$ g Pb/L in the 250 mL standing sample as a guide for remedial action. Further, visual inspection of the facility suggested that the sources of high lead concentrations were brass faucets and valves, and lead-based solder. Therefore, sequential samples were also taken at several locations. Sequential sampling involved taking a 250 mL sample from the site after a 10 minute flush and a sequence of two 60 mL samples and ten 125 samples following a 24 hour standing time. The samples were analyzed for lead, copper, and zinc.

Results from the DWRD sampling confirmed high lead levels in the drinking water at many locations in the building. Once again the levels were inconsistent from past results at the same site. Results from the sequential sampling typically showed high lead levels in the first two samples falling off to levels observed in flushed samples then peaking again at about the fifth or sixth sample in the sequence (example shown in Figure 1). Profiles for zinc, a major component of brass, typically showed the highest levels in the first and second samples, dropping rapidly to background levels. These test results suggested that the sources of lead were brass fixtures and lead-tin solder.

Visual inspection of the faucet, faucet connection, and plumbing material beneath sinks leading up to the faucet in each location sampled showed that the basic plumbing was generally the same for laboratories in the building. Typical cold water plumbing in the laboratories (Figure 2) had several potential sources of lead; solder joints, a brass bolt, and brass faucet. Slight differences in the number of solder joints existed among rooms.

## **Potential solutions**

Buildings, such as hospitals, office complexes, apartments, and schools have the same plumbing materials used in homes (e.g. copper pipes, brass faucets and fixtures, solder joints, etc..). Publicized problems related to high lead and/or copper in the tap water may likely have a significant negative impact on occupancy and may raise political issues. While plumbing systems in buildings are typically larger and more complicated in design than a home, water quality parameters that affect the solubility of lead and copper and treatment strategies to reduce the metal levels are the same. Issues that could complicate treatment in a building are more related to engineering, and flow and usage patterns of the system.

Based on the sampling results, DWRD concluded that the primary sources of lead were the solder joints and the brass faucets. To reduce lead concentrations in the drinking water, three options were provided: (1) remove all soldered copper plumbing, (2) install point-of-use (POU) devices at the taps, or (3) install a chemical treatment system. Removing the copper plumbing would eliminate the lead leached from the solder but not the lead from the brass fixtures. Secondly, such an option would be expensive, inconvenient, and time consuming. The disadvantages of POU devices were the cost of purchasing and installing devices at hundreds of faucet sites, maintenance, and the need for constant monitoring. Additionally, in-line systems would not solve the faucet lead problem. The installation of a chemical treatment system raised many questions by the employees. The major concern expressed was that adding chemicals containing zinc, silicate, or phosphate to the water supply could potentially impact their research studies.

## Water usage study

Building water was supplied by the Patuxent Water Treatment Facility, Patuxent, Maryland. The source water is river water treated by alum coagulation. Water quality parameters as measured at the treatment facility are shown in Table 1. The water quality would normally be considered relatively non-aggressive, however, testing results indicated aggressiveness towards new lead surfaces.

New plumbing systems are more susceptible to corrosion attack than older systems. Flux from the installation of solder joints, and oils and residues from the manufacturing of copper pipe and brass plumbing components can promote corrosion of the metal surfaces they contact. Metal particulate debris left on plumbing materials following manufacturing and installation practices can dislodge during system usage, contributing to tap water lead and copper levels. However, with time and usage or system "aging", these materials will be reduced or removed through dissolution and physical mechanisms. In addition, water usage will enhance the development of protective superficial and passivating films on the plumbing material, reducing metal diffusion from the metal surface. Therefore, the first proposal made by the DWRD was to evaluate the impact of water usage on metal levels throughout the new building's water supply. The DWRD designed a water usage study as well as offered to perform the necessary water sample analysis at the USEPA's Andrew W. Breidenbach Environmental Research Center (AWBERC), Cincinnati, Ohio.

The building plumbing system is split into 2 distinct halves, the "laboratory" and the "animal" sections. The laboratory section was designed for general lab experimentation while the animal section was designed as an isolation wing to prevent escape of biohazards to the outside. Based upon accessibility and plumbing considerations, the laboratory section was selected for the water usage study. The laboratory section was comprised of four levels (ground,  $1^{n}$ ,  $2^{nd}$ , and  $3^{nd}$  floors). Conveniently, the water lines feeding each floor could be isolated from the rest of the building. Each floor was comprised of two wings, each consisting of 9 rooms or laboratories, each with at least 1 faucet, that can be further isolated.

Two wings were selected for the water usage study; one on the ground floor and one on the third floor. A contractor was hired to supply a technician to turn on and off the faucets in the rooms. Flow meters were installed at the front of each wing to monitor water usage.

Beginning in early May, 1991, all faucets in nine laboratory rooms on each wing were opened for a total of 2 hours per day, 5 days per week. Faucets were opened four times a day for 1/2 hour with approximately 1-1/2 hour stagnation time between flow periods. Flow rate was approximately 1 L/min.

#### Sampling

Initially, sampling of the faucets was performed on every Tuesday and Friday of each week. Generally each room had two faucets, although some had one or three faucets. As a rule, one 250 mL sample was collected from the faucet furthest from the utility chase in each room prior to the first flow period on the specified days. This guaranteed at least a 12 hour standing time. The samples were preserved with ultrapure nitric acid (0.15% v/v) and immediately sent back to DWRD in Cincinnati, Ohio for analysis. Tuesday's samples were analyzed for lead, copper, and zinc, and Friday's samples were analyzed for lead only. Flame and graphite furnace methodologies were used for metals analysis. Friday sampling was eliminated in mid-October, 1991 to reduce laboratory workload after starting the chemical treatment portion of the study.

After approximately 100 days, two "flushed" 250 mL samples from two rooms of each wing were taken during the first flush period on Tuesdays. One sample was preserved with nitric acid (0.15% v/v) and analyzed for lead, copper, zinc, iron, calcium, potassium, and magnesium. The other sample was not preserved and was analyzed for alkalinity, phosphate, silicate, ammonia, sulfate, nitrate, and chloride. In addition, a hand held pH meter was used to monitor pH daily in the field.

### Chemical treatment study

By late Oc:ober, 1991, six month test data showed that although the lead levels had decreased slightly, the levels were still significantly above the desired maximum level of 20  $\mu$ g/L. At that time, a decision was made to evaluate chemical treatment. Based upon water quality and plumbing material considerations, three treatment chemicals were proposed for the study; (1) a 1:3 Zn:PO<sub>4</sub> (as mg/L) ratio zinc orthophosphate formulation, (2) "generic" alkali metal orthophosphate formulation, and (3) type "N" sodium silicate. Three different building wings, one on the 1" floor and two on the 2<sup>nd</sup> floor, were chosen to evaluate the corrosion inhibitors.

### Chemical feed system

A relatively simple, low maintenance, and inexpensive chemical feed system was designed and installed in the utility chase of each test wing. The feed system, shown in Figure 3, consisted of covered 100 gallon Nalgene<sup>®</sup> feed tank with mechanical stirrer. A Milton Roy  $A7^{1}$  feed metering pump was installed on the cover of the tank to feed chemical inhibitor to the existing 1-1/4" cold water line. The rate at which the chemical was fed was set by a Milton Roy RFP Series programmable flowmeter/pulsar apparatus. An in-line static mixer was included to insure sufficient chemical mixing. Chemical feed settings were adjusted to deliver approximately  $3.0 \text{ mg/L PO}_4^{-3}$  in the orthophosphate wings and

30 mg/L SiO<sub>2</sub> in the silicate wing. The materials cost for each feed system was approximately \$1300.

#### Feed chemicals

A general description of the treatment chemicals used in the study is presented in Table 2.

Sodium silicate solution was supplied by the PQ Corporation, Chester, PA. N-type sodium silicate with a 1:3.2 ratio of SiO<sub>2</sub>/Na<sub>2</sub>O was used. The solution contained 28.7% SiO<sub>2</sub>. The manufacturer recommended using a dose of 24 mg/L SiO<sub>2</sub> in the system during the first 30-90 days, falling back to a maintenance dose of 4-8 mg/L SiO<sub>2</sub>. Because of a lack of field data on the use of silicates for corrosion control and an understanding of corrosion control mechanisms by which silicates reduce lead and copper levels, a higher start-up dose of 30 mg/L was used. After approximately 70 days, the dose was decreased to a maintenance dose of 15 mg/L. Dosage recommendations were derived from research described in AWWARF's (1985) Internal Corrosion of Water Distribution Systems.<sup>3</sup> The sodium silicate feed tank was covered because sodium silicate exposed to the atmosphere becomes viscous, which may lead to clogging problems within the feed system. The solution is very basic, and would increase the pH of the treated water significantly because of the source water's limited buffering ability at the high SiO<sub>2</sub> level. The concentrated solution was diluted 1:2 with distilled water in the storage tank to accurately meter the basic feed solution.

The "zinc" orthophosphate formulation, product name SLI-939, was supplied by Shannon Chemical Corporation, Malvern, PA.. Commonly named zinc orthophosphoric acid, it's chemical formula is proprietary. However, it was identified by the manufacturer to contain 8% zinc and 24% phosphate as  $PO_4^{3}$  or a zinc to phosphate ratio of 1:3. Based upon manufacturer recommended dosage, a desire to quickly minimize lead solubility, previous experience<sup>4-7</sup>, and water quality, the dosage applied was 3 mg/L as  $PO_4^{3}$ . The concentrated solution was diluted 1:30 with distilled water in the storage tank.

The generic orthophosphate, product name SLI-1226, was also supplied by Shannon Chemical Corporation, Malvern, PA.. SLI-1226 is a proprietary blend of alkali metal orthophosphate salts in acidic solution and does not contain polyphosphate, silicate, or zinc. The orthophosphate content was 36% phosphate as  $PO_4^{3}$ . The dose used was also 3 mg/L as  $PO_4^{3}$  for the same reasons given previously and the solution was diluted 1:30 with distilled water in the storage tank.

# Sampling

On November 25, 1991, the chemical inhibitor test program began. All faucets in 9 laboratory rooms on each test wing were opened for a total of 2 hours per day, 5 days per week. Faucets were opened four times a day for 1/2 hour with approximately 1-1/2 hour stagnation time between flow periods. Flow

<sup>&</sup>lt;sup>\*</sup> Milton Roy, Acton, MA

rate was approximately 1 L/min. This protocol was identical to that followed in the water usage study.

Samples were take in the same manner as in the water usage study. Additionally, an unpreserved 125 mL flushed sample was taken from the first (closest to the chemical feed system) and the last room (farthest from the chemical feed system) in each wing to verify chemical dosing on Tuesday and Friday. These samples were analyzed for silicate or phosphate only.

The pH of the water was monitored daily on site using a hand held pH meter. Portable test kits were used on site to monitor consistency of phosphate or silicate dosing in the first and last room in each wing on a daily basis.

# Analytical methods and reagents

.

Water samples analyzed for metals were preserved on site by adding 0.15% v/v ultra pure reagent grade HNO<sub>3</sub><sup>+</sup> in accordance with EPA recommendations for preserving metals in drinking water samples.<sup>8</sup>

The analytical techniques used in this study are listed in Table 3. Method detection limits are also presented. Lead was analyzed with a Perkin-Elmer model 4000 atomic absorption spectrophotometer<sup>11</sup> equipped with a model HGA-400 graphite furnace and model AS40 autosampler. All other metal determinations were made on a Perkin-Elmer model 5000 flame atomic absorption spectrophotometer and model AS50 autosampler.

Alkalinity, chloride, and sulfate were measured with a Metrohm E-636 titroprocessor<sup>+++</sup>. Nitrate, phosphate, and silicate were measured on a Alpkem RFA/2 autoanalyzer<sup>++++</sup>.

Silica and phosphate determinations were made on site using a Hach<sup>11111</sup> SI-5 and a Hach PO-19A test kits, respectively. Sample pH was measured with a Cole-Parmer<sup>111111</sup> model 5985-75 pH meter and accompanying electrode.

Particulate material was analyzed with a Link EXL Energy Dispersive X-Ray System (EDXA)<sup>1111111</sup> mounted on a JEOL 5300 Scanning Electron Microscope (SEM)<sup>11111111</sup> at 30 kV.

# Results

# Water Usage study

The water usage study began on May 1, 1991 and was terminated on February 4, 1992, lasting about 9 months. During that time, nearly 260,000 gallons of water (1500 gallons/day) and 106,000 gallons of water (600 gallons/day) were flushed through the ground and third floor wings, respectively. Water usage was greater on the ground floor wing because, as it was later discovered, two of the nine rooms on the wing had been used prior to and during the study by the building's janitorial and maintenance employees. In addition, towards the end of the study, employees began to gradually move into the building. The nature of their work required the employees to lock unattended laboratories. During the final month of the study, it was often impossible to enter locked laboratories and proceed with water usage study.

Results from the usage study indicated no apparent reduction in lead levels with water usage over the time period of this study. Variability in the data because of frequent lead spikes tended to statistically suggest that water usage had no effect on lead levels. For example, a room would have low lead values (<20  $\mu g/L$ ) during a sequence of daily sampling, then a few very high values (>50  $\mu g/L$ ). The sporadic occurrence of high lead values occurred regularly among nearly all of the rooms sampled in the usage study. The lead distributions for the rooms on the third floor wing is shown in Figure 4. Some lead spikes were greater than 1000  $\mu g/L$ . Two rooms on the ground floor (Figure 5) appeared to be exceptions. As mentioned, these two rooms were used prior and during the study by the janitorial employees. The additional water usage probably contributed to the lower overall lead concentrations and reduced the occurrence of sporadic lead spikes, suggesting a very long time of usage may reduce the problem after all.

The occasional unusually high lead levels in samples, and a general non-Gaussian pattern of the lead levels, are in accord with many previous laboratory and field studies of lead corrosion.<sup>4,5,9-16</sup> In addition to occurring as dissolved aqueous ions and complexes, lead can be present as, or associated with, various colloids or particulates. These solids can originate as non-adherent corrosion deposits, eroded pieces of plumbing material, or be present in the background water in the building or municipal distribution system. Many kinds of particles have been shown to be effective scavengers of lead in natural and potable waters. Notable among the solids having a high affinity for sorbing or incorporating lead are hydrous iron oxides<sup>12-14,17</sup>, humic substance colloids (with and without associated iron)<sup>13,16,18</sup>, calcite particles<sup>19</sup>, and various stream sediments.<sup>20</sup> In several cases, the sorption of lead on particle surfaces can be explained by either a "surface complexation" approach<sup>21</sup> or by a cation-exchange process. In either of these two cases, lead should usually be readily removed from the particles after sample acidification. Thus, the lead originally associated with the particles would be detected in the same manner as dissolved lead by conventional graphite furnace

<sup>&</sup>lt;sup>†</sup> Ultrex, J.T. Baker Chemical Comp., Phillipsburg, NJ

<sup>&</sup>lt;sup>11</sup> Perkin-Elmer Corp., Norwalk, CT

<sup>&</sup>lt;sup>+++</sup> Metrohm, Switzerland

<sup>&</sup>quot;" Alpkem Corp., Wilsonville, OR

<sup>&</sup>quot;"" Hach, Loveland, CO

<sup>&</sup>quot;"" Cole-Parmer, Chicago, IL

<sup>&#</sup>x27;''''' Link Analytical, Madison, WI

<sup>&</sup>quot;""" J.E.O.L., Peabody, Mass

atomic absorption spectroscopy (GFAAS) or inductively-coupled plasma spectroscopy (ICP).

Often, visual examination of the acid-preserved (0.15% HNO<sub>3</sub>) water samples, showing abnormally high lead concentrations, revealed that metal particulates were present at the bottom of the sample. On several occasions, when samples showing unusually high lead concentrations were reanalyzed on later days, the concentrations tended to be inconsistent, and slightly increased to some eventually-consistent level. Originally, the spikes were thought to be an instrumental problem, before the samples were more thoroughly examined. The observations were later found to coincide reasonably with occasions when samples were analyzed within several hours of receipt from the field site, reflecting 1-2 days of total clapsed time after sampling and immediate acidifications.

Ten samples (from rooms in the two usage wings) analyzed with high lead concentrations and identified as containing particulates were filtered through 0.2  $\mu$ m polycarbonate filters. The particles retained on the filters were examined by an energy-dispersive X-ray analysis (EDXA) system attached to a scanning electron microscope (SEM) in an attempt to characterize the source of lead. In all cases, the individual particles examined showed EDXA elemental spectra containing tin, but lead or lead-containing particulates were not found. When tin particles were found, high lead levels were likely caused by the corrosion of the lead-tin solder. Lead or lead-containing particulates were likely to coexist with tin or tin-containing particles in the original samples. Therefore, preferential dissolution of lead phases by the acid preservative was strongly implicated. These observations have been discussed in detail in another paper.<sup>22</sup>

Copper levels did not follow the same sporadic peak trend set by lead levels (shown in Figure 6 for the third floor). There were a few occasional variations in copper levels, however, typically several rooms experienced these variations or small peaks on the same day. This suggests the influence of water quality parameter changes (e.g. pH and chlorine residual) rather than particulate material. Previous experience has demonstrated that copper can be very sensitive to small pH and chlorine fluctuations.<sup>23</sup> Visually, there did not appear an obvious overall trend, although it could be argued that copper levels increased slightly over time. Copper levels were relatively low, nearly all were less than the 1.3 mg/L action level set by the Lead and Copper Rule<sup>24.25</sup>. Although the Rule specifies 1 L samples, it stands to reason that 1 L samples taken from the rooms would remain below 1.3 mg/L.

Zinc levels more closely followed the leaching patterns described by copper levels than lead levels (shown in Figure 6 for the third floor). There were a few more random zinc peaks, some of which could be more related to particulate material than water quality changes. Two zinc spikes, in room 3301 (at approximately 100 days) and in room 3329 (at approximately 50 days), also have corresponding lead peaks on those days. This suggests that brass particulate, possibly coming from mechanical or hydraulic abrasion of brass surfaces in the faucet, was present in those samples. However, with only two standout cases of zinc peaks, brass does not appear to be a significant source of lead spikes.

Background water quality measurements represented by flushed samples and field pH measurements were not initiated until late into the usage study as previously mentioned. Table 4 shows pH and other water quality parameters of the background water in flushed samples taken from the treated wings. Background water quality agreed well with water quality leaving the Patuxent Water Filtration Plant (Table 1).

# Chemical treatment

Chemical treatment began on November 25, 1991 and ended on April 14, 1992, lasting just over 4 months. Prior to the start of chemical treatment, one set of "baseline" samples was taken from the faucets in the 3 wings (shown in Table 5). Only one set could be collected because the immediate necessity of the chemical treatment evaluation limited the length of time such an evaluation could take place. The absence of a statistically viable baseline precluded the later application of objective statistical procedures for objective evaluation of comparative treatment performance. However, based upon building construction records, plumbing materials used, visual examination of the plumbing, baseline results, and previous sampling results, it is reasonable to assume that the wings used in the treatment study would behave similarly to those wings in the usage study with respect to metal leaching trends. Also, instrumental malfunctions associated with the silicate and phosphate analysis conducted by the EPA resulted in eliminating nearly all phosphate and silicate laboratory analysis. Therefore, silicate and phosphate concentrations monitored in the field were the most reliable determinations of chemical feed doses.

#### Zinc orthophosphate

During the study period, 101,277 gallons of water was passed through the plumbing of the nine rooms in the zinc orthophosphate test wing. Phosphate levels were maintained at approximately 3.3 mg  $PO_4^{3/1}L$  (see Table 6). It appeared that phosphate deposition between the first room (room closest to the chemical feed system) and last room in the wing accounted for a 0.2 mg/L phosphate concentration drop. Zinc orthophosphate reduced the pH by about 0.5 pH units, from approximately 7.7 to 7.2. Other than zinc, which increased by approximately 1.25 mg/L, no other water quality parameter was notably changed from background water quality using zinc orthophosphate inhibitor (see Table 4).

Zinc orthophosphate effectively reduced lead levels in the wing. Lead levels among the majority of rooms in the wing dropped rapidly and stabilized at less than 5  $\mu$ g/L by 80 days (shown in Figure 8). In addition, the occurrence of random lead spikes among samples was nearly eliminated.

Zinc orthophosphate addition reduced copper levels. As with lead, copper levels appeared to stabilize after about 80 days of treatment. Copper levels remained below 0.15 mg/L after stabilization.

Logically, zinc levels in the water treated with zinc orthophosphate became much higher than in water treated with other treatment chemicals. Zinc levels increased approximately 1.25 mg/L. Zinc concentrations correspond well to phosphate concentrations in that the ratio of zinc to phosphate was approximately 1:3; the mean zinc concentration was 1.3 mg/L and the mean phosphate concentration was 3.3 mg  $PO_4^{-3}/L$  (background zinc and phosphate levels were insignificant).

# Generic orthophosphate

Over the study period, 92,676 gallons of water was passed through the plumbing in the eight rooms of the calcium orthophosphate test wing. Phosphate levels were maintained at approximately 3.3 mg  $PO_4^{3}/L$  (see Table 6). It appeared that phosphate deposition between the first room (room closest to the chemical feed system) and last room in the wing accounted for about a 0.1 mg/L phosphate concentration drop. Orthophosphate dosing reduced the pH of the rooms in the 2300 wing by about 0.4 pH units, from approximately 7.7 to 7.3. No other water quality parameter was notably changed from background water quality using calcium orthophosphate (see Table 4).

Lead levels in the water were effectively reduced by the orthophosphate addition and appeared to drop and stabilize more rapidly with generic orthophosphate than with zinc orthophosphate. Lead levels generally dropped to and stabilized at about 5  $\mu$ g/L after only 25 days (shown in Figure 9). However, there were exceptions. Room 2305 took almost 100 days to stabilize at 10  $\mu$ g/L. This was probably related to the plumbing in the room; perhaps there were more solder joints used in the room's plumbing. Room 2303 stabilized after 25 days but increased dramatically after 50 days. This behavior is inconsistent with solubility behavior and normal orthophosphate passivation behavior, suggesting perhaps the plumbing was disturbed in some way. The sporadic occurrence of lead pcaks was generally reduced among rooms.

Copper levels were also reduced and appeared to stabilize after about 50 days at less than 0.2 mg/L by orthophosphate dosage. There were only a few small random copper peaks of almost 1 mg/L. The appearance of the peaks is consistent with behavior of particulate material, however lack of verifiability of odd trends is a problem with remote field sampling. There were no corresponding lead or zinc peaks which might suggest the source of copper to be copper pipe particles rather than brass.

Zinc levels were reduced almost immediately by the addition of the generic orthophosphate. Zinc levels were maintained below 0.2 mg/L. No significant peaks appeared in the data.

## Sodium silicate

Over the study period, 96,476 gallons of water was passed through the plumbing in the nine rooms of the sodium silicate test wing. Silicate dose started at about 32 mg SiO<sub>2</sub>/L, and after 71 days was dropped to a maintenance dose of 16 mg SiO<sub>2</sub>/L (see Table 6). It appeared that silicate deposition between the first (room closest to the chemical feed system) and last room in the wing accounted for about 1.0 mg/L silicate concentration drop for both doses. The addition of sodium silicate at the start-up dose raised the pH of the treated water by more than 1.5 pH units, to about 9.5. The pH dropped to a range of 8.8-9.1 after reducing to the maintenance silicate dose. Sodium levels resulting from sodium silicate addition increased sodium levels by only about 4 mg/L over background.

Lead levels dropped rather rapidly, stabilizing after about 25 days at approximately 10  $\mu$ g/L (shown in Figure 10). In addition, the occurrence of sporadic lead peaks was greatly reduced. An exception was room 2408 that stabilized at about 25  $\mu$ g/L. The higher lead levels may be an artifact of the plumbing; perhaps there were more exposed solder joints or the workmanship related to the soldering was poor. Also, room 2416 was very peculiar in that the lead levels continuously climbed. This is difficult to explain and would require closer examination of the plumbing in that particular room for a full analysis. Dropping to the maintenance silicate dose appeared to make no impact on lead levels. As mentioned, the silicate doses used were greater than manufacturer recommended doses.

Copper was almost immediately reduced (shown in Figure 11). Sodium silicate appeared to be the best of the chemical inhibitors at reducing copper levels, maintaining levels below 0.07 mg/L. Also, there were no sporadic copper peaks except for one exception, room 2402. Copper levels appeared to stabilize at about 0.25 mg/L, far above normal solubility levels of any oxide, hydroxide, or basic carbonate solids of  $Cu^{2+}$ . Lead values in this room however, were not noticeably different than the other rooms in the wing.

Zinc levels were also reduced almost immediately by sodium silicate addition. Levels stayed below 0.05 mg/L with the exception of room 2402 which was sporadic and about 0.5 mg/L. This room also had high copper levels which might indicate excess brass plumbing or high erosion of plumbing material.

# Discussion

The results of the water usage study indicated that high lead levels in water from a new building may take a long time to drop under conditions of "normal" water usage; in the case of the agency building, greater that 8 months. Lead levels of the water samples taken bi-weekly showed overall high and inconsistent lead levels with the occurrence of random lead spikes. Sources of lead were identified as solder joints and brass fixtures. Extraordinary high lead concentrations or spikes in that data were probably due to lead-containing

particulate material. This type of occurrence is not uncommon. At this building, a major contributor to the particulate material causing the erratic lead levels was found to be lead-tin solder. Lead containing particles could also be coming from brass, however solder was discovered indirectly to be the cause because of the insoluble tin that was sometimes left in the samples with high lead concentrations.

Zinc orthophosphate, generic orthophosphate, and sodium silicate treatment effectively and quickly reduced lead levels and the occurrence of lead spikes. All treatments also reduced copper concentrations. Differences in treatment performances may strictly be due to variations in plumbing configurations such as the number of solder joints or the workmanship of those joints. Results are strictly based on observations and would be difficult to prove statistically with any certainty. Every wing had at least one room in it that was an exception to these general rules. Exceptions may be due to differences in plumbing configuration or usage patterns. However, all of the treatment chemicals reduced lead in the water to acceptable levels.

To further reinforce the proceeding observations, box plots were constructed for each treatment wing (Figures 12-15). Box plots offer the ability to visually and statistically describe data sets. Box plots were used to identify general characteristics of the distribution of lead levels in each wing over time. In other words, the effect water usage and corrosion inhibitors have on the lead distributions of all the rooms of a wing can be tracked. Each box shows various distribution percentiles and the mean of the lead values for the 8 or 9 rooms in the wing for each day. Plots clearly show that lead levels and the sporadicness are reduced (percentile ranges are reduced) by all chemical treatments.

In the absence of a long enough period of pre-treatment monitoring to establish a true baseline, statistical tests could not be made to quantify relative differences caused by the treatments. The variability among rooms in the usage study as well as within treatment wings also argues that more sites would probably be needed for controlled evaluations.

### Conclusions

The following conclusions from the corrosion control study conducted at the agency facility can be made:

1. General water usage, as described by the test protocol in this paper, did not appear to reduce lead levels in the drinking water during the duration of the study. Results indicated that lead reductions by continuous water usage in a building may take more than 8 months and probably years.

2. Water samples taken during the water usage study showed inconsistent lead levels among daily samples, often with sporadic lead concentration spikes. Particulate material, most likely Pb:Sn solder, was the major contributor of the larger random lead spikes.

3. A simple, economic, low maintenance chemical inhibitor feed system can be designed and installed to add inhibitor to an existing building plumbing system that will produce consistent application of a variety of liquid chemical inhibitors.

4. Zinc orthophosphate, alkali metal orthophosphate, and sodium silicate corrosion control inhibitors all reduced lead concentrations to acceptable levels. Although copper was not an issue, they all reduced copper levels as well, at the dosages and respective pH ranges employed.

5. Zinc orthophosphate, alkali metal orthophosphate, and sodium silicate corrosion control inhibitors all reduced and nearly eliminated the occurrence of random lead spikes in daily monitoring

The success of this study was based on theoretical considerations and previous knowledge and experience of using silicate- and phosphate- based corrosion inhibitors in drinking water systems. An understanding of the water quality conditions (ie. pH, DIC, etc.) that favor their usage and proper dosage rates is essential if success in reducing lead and copper solubility is to be achieved.

# Acknowledgements

The authors would like to thank the following members of the USEPA, DWRD, IPCB staff for their contributions: Keith Kelty, James Doerger, James Caldwell, and Louis Trombly for performing the many water analysis's, and Herb Braxton for graphical assistance and water analysis. We would also like to thank Greg George, Roger Rickabaugh, Steve Harmon, and John Dammann of Technology Applications Incorporated for their technical assistance.

## Disclaimer

Mention of specific trade names or instrument models is for explanatory purposes only, and does not constitute an endorsement by the U.S. Environmental Protection Agency.

### References

1. Office of Water. "Suggested Sampling Procedures to Determine Lead in Buildings Other Than Single Family Homes", U.S. Environ. Protection Agency, Washington, D.C. (June, 1988).

2. Office of Water. "Lead in School Drinking Water", EPA-570/9-89-001. U.S. Environ. Protection Agency, Washington, D.C. (Jan., 1989).

3. American Water Works Association Research Foundation and DVGW Forschungsstelle. Internal Corrosion of Water Distribution Systems. A, Denver, CO. (1985).

4. Schock, M. R. Understanding Corrosion Control Strategies for Lead. Jour. AWWA, 81:7:88 (1989).

5. Colling, J.H., Whincup, P.A.E., and Hayes, C.R. The Measurement of Plumbosolvency Propensity to Guide the Control of Lead in Tap Waters. *Jour. Inst. Water and Environ. Mgmt.*, 1(3):263-269 (1987).

6. Gregory, R., and Jackson, P.J. Central Water Treatment to Reduce Lead Solubility. In *Proc. AWWA Annual Conf.*, Dallas, TX. A, Denver, CO. (1984).

7. American Water Works Association Research Foundation. Lead Control Strategies. AWWA, Denver, CO. (1990).

8. Environmental Monitoring and Support Laboratory. Handbook for Analytical Quality Control in Water and Wastewater Laboratories. EPA-600/4-79-019. U.S. Environ. Protection Agency, Cincinnati, OH (1979).

9. Schock, M. R. Causes of Temporal Variability of Lead in Domestic Plumbing Systems. Environ. Monit. & Assessment, 15:59, (1990).

10. Schock, M. R. et. al. The Significance of Sources of Temporal Variability of Lead in Corrosion Evaluation and Monitoring Program Design. Proc. AWWA WQTC, St. Louis, MO (1988).

11. Britton, A. & Richards, W. N. Factors Influencing Plumbosolvency in Scotland. Jour. Inst. Water Engr. & Scientists, 35:4:349 (1981).

12. Hulsmann, A. D. Particulate Lead in Water Supplies. Jour. Inst. Water Engnr. & Mgmt., 4:2:19 (1990).

13. Breach, R. A., et. al. A Systematic Approach to Minimizing Lead Levels at Consumers Taps. Proc. AWWA Annual Conf., Philadelphia, PA (1991). 14. Harrison, R. M. & Laxen D. P. Physicochemical Speciation of Lead in Drinking Water. *Nature*, 286:791 (Aug. 1980).

15. de Mora, S. J. & Harrison, R. M. Lead in Tap Water: Contamination and Chemistry. *Chem. Britain*, 20:900, (1984).

16. de Mora, S. J. et. al. The Effect of Water Treatment on the Speciation and Concentration of Lead in Domestic Tap Water Derived from a Soft Upland Source. *Water Res.*, 21:1:83, (1987).

17. de Lurdes Simões Gonçalves, M., et. al. Voltammetric Methods for Distinguishing between Dissolved and Particulate Metal Ion Concentrations in the Presence of Hydrous Oxides. A Case Study on Lead(II). Environ. Sci. & Technol., 19:141 (1985).

18. Salim, R. Adsorption of Lead on the Suspended Particles of River Water. *Water. Res.*, 17:4:423 (1983).

19. Wouters, L. C. et. al. Discrimination between Coprecipitated and Adsorbed Lead on Individual Calcite Particles Using Laser Microprobe Mass Analysis. Anal. Chem., 60:2218 (1988).

20. Hem, J. D. Geochemical Controls on Lead Concentrations in Stream Water and Sediments. Geochim. Cosmochim. Acta, 40:599 (1976).

21. Osaki, S. et. al. Adsorption of Fe(III), Co(II) and Zn(II) onto Particulates in Fresh Waters on the Basis of the Surface Complexation Model. II. Stabilities of Metal Species Dissolved in Fresh Waters. Sci. Tot. Environ., 99:115 (1990).

22. Lytle, D.A., Schock, M.R., Dues, N.R., and Clark, P.J. Investigating the Preferential Dissolution of Lead From Solder Particulates. *Jour. AWWA*, 7:85:104-110 (July 1993).

23. Lytle, D.A., Schock, M.R., and Tackett, S. Metal Corrosion Coupon Study Contamination, Design, and Interpretation Problems. In *Proc. AWWA Water Quality Technology Conf.*, Toronto, Ontario, Canada (1992).

24. Lead and Copper. Final Rule. Fed. Reg., 56:26460 (June 7, 1991).

25. Lead and Copper. Final Rule Correction. Fed. Reg., 56:135:321132 (July 15, 1991).

			Minimum	Махіти	5
	Unit of	Ycarly	Monthly	Month	Y
Parameter	Measurc	Average	Average	Averag	0
Physical Allesterity	me/l	32	25		39
Mitanuly Discrimed Colide Total	ine/L	105	86	-	14
I Jissoived Julius, 1 Julius	T/am	52	46		26
	Units	8.0	7.5	~	3.2
pro Turbidity	NTU	0.16	0.10	Ċ	10
Metals					
Aluminum	mg/L	0:030	0000	0.0	44
Arconic	T/am	0.001	0.000	0.0	S
Bartim	J/am	0.04	0.02	0	01
Cadmium	me/L	0.001	000	0.0	ŝ
Calcium a	me/L	17.4	13.1	2	6.0
Chromium	mg/L	0.001	00.00	0.0	80
Conner	2/300	0.012	0.00	00	8
linn	ng/L	0.008	00.00	0.0	013
Lead	_√gœ	0:001	00.0	0.0	Ő
Magnesium	ng/L	3.4	~	2	3.6
Manganese	mg/L	0.002	00.0	1	§
Mercury	mg/L	0.0000	0.00	0.0	ŝ
Potassium	J/gm	2.6	2		3.2
Selenium	J/gm	0.0004	0.00	0.0	010
Silicon	mg/L	1.7	Ö	6	2.6
Silver	ng/L	100.0	0.00	0	013
Sodium	mg/L	6.2	Š	4	7.0
Zinc	1/acr	0.003	0.0	o ç	015
lacessice					
Roron	- mg/L	0.01	0.0	2	0.03
Carbon Dioxide	mg/L	1.7	0	9	4.1
Chloride		14.4	10	.2	18.0
Chlorine	mg/L	1.6	-	s.	1.9
Discoved Orvern	mg/L	9.6	v.	80	14.7
Fluoride	ng/L	06.0	0.	\$	0.95
Nitrate as Nitrogen	mg/L	1.40	0	94	2.10
Nitrite as Nitrogen	ng/L	0.003	0.0	8	10
Phoenhorous	mg/L	0.02	0.	8	0.12
Sulfate		11.8	æ	5.7	15.0
			. Thidin 1)	nite	
• Notes:		cpiterouncuts Giliorams ner	Liter (Parts	per Billio	â
		JŸ-mi			

· Effective pH is heavily dependent upon dosage. These dosages are unlikely to be effective below a pH of approximately 7.5.

Sodium mubo2	Na;:nSiO,, where n is typically 2–3.2. Chemical is a liquad containing 28.7% SiO;,	Start up dose: 24–30 աջ/L as SiO <sub>2</sub> (for 3 monthes) Maintainance dose: 10– 16 ոք/L as SiO <sub>3</sub>	0.7 <	Raises (Depending on buffering intensity)	AWWA :(72V) A932U
ອາຣກ່ຽວກ່ຽວກ່າວ "ຈາາອາອານີ"	Typically a blend of Na and K orthophosphate salts (c.g. Na <sub>H</sub> 2O4) in an acidic solution. Contains 36% PO <sub>4</sub> .	2 - 4 mg/L 22 PO'}-	.5.2-9	Lowers slightly (Depending on buffering intensity)	awwa :(Asp) aged
sisriqzorkqort10 °oni∑"	Typically a Mend of ZnSO, and VaH, PO, in an acidic solution. Contains 24% PO,	2 - ¢ m8\J ss PO <sup>4</sup> 2-	.5.6-0	Lowers slightly (Depending on buffering intensity)	USEPA (NSF): AWWA
Chemical	leaimart.D formulation	gose Recommended	Effective Pq	Effect on pH	Vpproval

Table 1. General characteristics of the treatment chemicals used in the building study.

Parameter	Method Number	Reference	Detection Limit (mg/L)
Calcium	7140	EPA*	1.0
Copper	7210	EPA'	0.02
Iron	7380	EPA <sup>4</sup>	0.05
Lead	7421	EPA	0.002
Magnesium	7450	EPA*	2.0
Manganese	7460	EPA"	0.01
Potassium	7610	EPA*	0.07
Sodium	7770	EPA'	0.75
Zinc	7950	EPA*	0.01
pН	9040	EPA*	
Chloride	9252	EPA*	1.0 (as CI)
Sulfate	9038	EPA*	7.0 (as SO)
Alkalinity	310.1	EPA	1.0 (as CaCO,)
Silicate	A303-S220-13	Alpkem <sup>c</sup>	0.4 (as SiO_)
Nitrate	A303-S173-00	Alpkem	0.02 (as N)
Phosphate	A303-S200-02	Alpkem <sup>c</sup>	0.1 (as PO)
Phosphate	8048	Hach <sup>d</sup>	•
Fotal chlorine	8167	Hach <sup>d</sup>	0.7 (as Cl.)
Free chlorine	8021	Hachd	0.7 (as Cl <sub>2</sub> )

Table 3. Analytical techniques used to measure water quality parameters.

a "Test Methods for Evaluating Solid Wastes." (SW 846) RREL, U.S. EPA, September, 1986

b. "Methods for Chemical Analysis of Water and Wastes" U.S. EPA, Revised 1983.

c "RFA/2 Methods Manual." Alpkeni, April, 1991.

d "Hach DR/2000 Spectrophotometer Methods Manual." Hach Company, Loveland, CO. (1992).

Treament	Statistic	Hq	Akalmıç	បី	G	5	Х	Å	۳ Z	NH3	NO3	£	PO4 <sup>3 -</sup>	SiOz	so4	5
WUR		Sim	mg causel			Ш М		mg/r				U8'L		mg/L		mg/L
Usage	Mean	7.69 <sup>a</sup>	30.8	18.3	19.3	0.05	2.4	4.0	6.5	< 0.05	1.43	< 2.0	< 0.1	6.4	12.5	< 0.1
)	Std. dev.	0.27	2.4	2.0	3.1	0.02	0.1	0.2	0.6	I	0.16	١	I	1.0	3.5	1
	Min.	40.7	26.8	16.1	15.6	< 0.02	2.4	3.7	5.5	ł	1.22	ł	I	5.1	8.6	۱
	Max.	8-37	7.60	21.3	25.4	0.07	2.5	4.3	<b>1</b> .7	ł	1.63	4.0	I	8.0	16.8	0.01
Sodium silicate	Mean	8.973	EU	17.5	na	0.03	2.5	4.0	10.2	50	na	< 2.0	ца 3	12.0•/16.3•• <sup>a</sup>	na	0.02
	Std. dev.	0.18	na	1.6	na	0.05	0.1	0.2	0.6	53	EU	١	na	5.9/0.8	Ш	0.06
	Min.	8.20	na	15.5	B.U	< 0.02	2.4	3.7	9.6	50	na	i	na	10.0/12.0	па	< 0.1
	Mar.	9.29	EU	21.6	na	0.16	2.6	4.4	11.0	na	na	١	ВП	39.0/17.0	50	0.25
Generic orthophosphate	Mean	7.26 <sup>a</sup>	En 13	17.4	na	0.03	2.5	3.9	6.6	50	na	< 2.0	3.3 <sup>a</sup>	Па	БЦ	0.02
	Std. dev.	<b>60</b> .0	na	1.8	na	0.04	0.1	0.2	1.1	ВП	<b>D</b> 3	ł	0.2	вn	U	0.04
	Min.	7.13	na	15.5	na	< 0.02	2.4	3.7	5.2	n3	ВЦ	١	3.0	па	na	< 0.1
	Mar.	7.51	00	21.6	50	0.16	2.6	4.4	9.2	пa	na	20	3.8	na	na	0.15
Zinc orthophosphate	Mean	7.22 <sup>a</sup>	5	17.3	na	0.03	2.5	3.9	6.8	na	na	< 2.0	3.4 <sup>a</sup>	50	ĉ	1.27
	Std. dev.	0.0	na	1.5	na	0.06	0.1	0.2	1.2	60	<b>П</b> 3	۱	0.2	5U	ũ	0.17
	Min.	7.07	na	15.5	Da	< 0.02	2.3	3.7	5.6	na	<b>na</b>	1	2.7	EU	č	0.0
	Max.	7.46	na	20.5	na	0.21	2.6	4.2	9.6	<b>6</b> 0	Eu	ł	3.5	<b>U</b> 3	ë	1.43
<ul> <li>Start-up dosage</li> <li>Mamtainance dosage</li> <li>Measurement made in</li> <li>na Not available</li> </ul>	the field															

Table 4. Background water quality in the building treatment wings.

					Roc	m Num	ber			
					Le	ead (µg/1	.)			
2400 Wing	Date	2402	2404	2406	2408	2410	2412	2414	2416	2418
(Sodium Silicate)	09/19/91	60.2	12.5	79.9	312.5	88.3	147.0	80.8	149.8	59.4
2300 Wing	Date	2301	2303	2305	2311	2313	2315	2319	2321	
(Generic Orthophosphate)	11/20/91	62.3	104.1	257.4	574.7	128.5	91.7	113.5	102.4	
1400 Wing	Date	1402	1404	1406	1408	1410	1412	1414	1416	1418
(Zinc Orthophosphate)	09/19/91	33.5	4203.0	69.3	599.0	68.3	88.7	44.2	140.7	

Table 5. Baseline lead levels in chemically treated building wings.

.

Table 6. Chemical duses in treatment wings based on field measurements.

E 1 1 20 / as PO 1 0,18 0 04	Generic onhophosphate         First         3.30 (as PO,)         0.17         0.04           Last         3.18         0.21         0.04	Sodium silicate         First         16.34 (as SiO <sub>2</sub> )         0.83         0.24           (maintenance dose)         Last         15.33         1.03         0.38	Sodium silicare         First         31.97 (as SiO <sub>2</sub> )         5.8.5         1.95           (start-up dose)         Last         31.06         4.2.8         1.45	Standard Mean deviauon 95% Cl Chemical Room (mg/L) (mg/L) (mg/L)
0.18 0.04	0.17 0.04 0.21 0.04	0.83 0.24 1.03 0.38	585 1.95 4.28 1.45	Standard deviation 95% C (mp/L) (mp/L)
0.18 0.04	0.17 0.04 0.21 0.04	0.83 0.24 1.03 0.38	5.85 1.95 4.28 1.45	tandard cvration 95% C (mp/L) (mp/L)
3.8	3.5 8	17 17	96 E	Minimum (ng/L)
19	30 27	10	10 12	Mຈນເກເພາ ( <del>ຫ</del> ຊ <sup>/</sup> L)

365





Figure 2. Typical cold water plumbing connection to brass faucet. Numbered locations represent visible soldered connections (drawing not to scale).





















Т



.

Figure 13. Box plot distribution of lead of rooms in the wing treated with zinc orthophosphate.

,



Figure 14. Box plot distribution of lead of rooms in the wing treated with alkali metal orthophosphate.



Figure 15. Box plot distribution of lead of rooms in the wing treated with sodium silicate.

TECHNICAL REPORT DATA (Please read Instructions on the reverse before comple								
1. REPORT NO. 2.	3.							
4. TITLE AND SUBTITLE	5. REPORT DATE							
A Systematic Study on the Control of Lead in	a New							
Building	6. PERFORMING OF	GANIZATION CODE						
7. AUTHOR(S)	8. PERFORMING OF	GANIZATION REPORT NO.						
Darren A. Lytle, Michael R. Schock & Thomas	J. Sorg							
9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEP	MENT NO.						
USEPA, DWRD, I&PCB, RREL 26 W. Martin Luther King Dr	11. CONTRACT/GR	ANT NO.						
Cincinnati, OH 45268								
12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPOR	AT AND PERIOD COVERED						
USEPA	Published	Paper						
26 W. Martin Luther King Dr								
Cincinnati, OH 45268	EPA/600/14	ł						
15. SUPPLEMENTARY NOTES 1994 AWWA Annual Confere NY, P:345-380	ence Proceedings, 6/18-23/	94, New York City,						
Project Officer = Darren Lytle (513)569-7432								
A new building was identified as having high lead levels in its drinking water.								
Through a detailed sampling protocol, the so plumbing fittings and fixtures, and Pb:Sn so lated sections of the building plumbing syst be reduced naturally with time by simply usi lead levels were not achieved following 8 mo performed to evaluate the effectiveness of 1 orthophosphate, calcium orthophosphate, and Three economic, simple, low maintenance chem installed in three different isolated section The chemicals were fed into the building sec showed that all inhibitors effectively and n	burces of lead were identi- older. A study was perfor- tem to determine if the lea- ing the water. Significan onths of water usage. A s- three chemical corrosion i sodium silicate, to reduc nical feed systems were de ons of the building's plum ctions for approximately 4 rapidly reduced lead and c	fied as brass med in two iso- ad levels could t reductions in econd study was nhibitors: zinc e the lead levels. signed and bing system. months. Results opper levels.						
17. KEY WORDS AND DO	CUMENT ANALYSIS	COSATI Field/Group						
a. DESCRIPTORS	D.IDENTIFIERS/OPEN ENDED TERMS	c, cosari ricu/oroup						
CORROSION CONTROL BUILDING CORROSION LEAD CORROSION COPPER CORROSION ORTHOPHOSPHATE BASED CORROSION INHIBITORS SILICATE BASED CORROSION INHIBITORS								
18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES 37						
	20. SECURITY CLASS (This page)	22. PRICE						
RELEASE TO PUBLIC								

,

ì

1