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Impact of advanced water conservation features and new copper pipe on rapid chloramine decay and microbial regrowth

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ABSTRACT

Taste and odor issues occurring in new buildings were attributed to rapid loss of chloramine residual, high levels of microbes in the potable water system, and high water age due to use of advanced water conservation devices. Laboratory experiments confirmed that chloramine could decay rapidly in the presence of new copper pipe, providing a possible explanation for the rapid disinfectant loss in the new buildings. Higher temperature and lower pH also accelerated the rate of chloramine decay in copper pipes. The reaction was slowed by the addition of phosphate inhibitor or aluminum, which presumably formed barriers between the pipe wall and the chloramine in the bulk water. Additional research is needed to better understand how to maintain high quality water in buildings while also conserving water.

1. Introduction

Monochloramine (NH_2Cl, chloramine) is usually highly persistent compared to free chlorine (HOCl, OCl^-) (Neden et al., 1992; Norton and LeChevallier, 1997) and is well-suited to maintain disinfectant residuals to consumers' taps. Chloramine also is often more effective than free chlorine in reducing heterotrophic plate counts (HPC) in potable water systems (Neden et al., 1992). These benefits have increased the popularity of chloramine disinfection among U.S. water utilities. Chloramine is thought to form fewer disinfection by-products (DBP) with natural organic matter (NOM) compared to free chlorine. However, more iodinated DBPs can form in the presence of iodine (Bougeard et al., 2010; Hua and Reckhow, 2007), and more nitrosamines form from reactions with nitrogen-containing compounds during chloramination (Mitch and Sedlak, 2002).

1.1. Sustainable construction and high water age

Sustainable construction reduces potable water use through the increased use of advanced water conservation features in buildings (e.g., metered and sensor faucets, low-flow showerheads and toilets, waterless urinals, and non-potable water use for toilet flushing). When potable water use in buildings is lowered without also reducing pipe diameters, stored water volumes (water residence time) markedly increases. Excessive residence time is known to cause microbial regrowth in water

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distribution systems regardless of the disinfectant type (USEPA, 2002). Regrowth is expected to be particularly acute in premise plumbing, which characteristically has a large pipe surface area-to-water volume ratio, regular periods of stagnation, and variable temperature and redox potentials (Table 1). Plumbing codes make every building a “dead end”, where water can stagnate in the pipes of conventional plumbing systems for hours or days before use and can form ecological niches for a wide diversity of microorganisms (Edwards, 2004; Edwards et al., 2003; National Research Council, 2006).

Potable water regulations historically have applied to main distribution systems but not building plumbing systems. The notable exception in the U.S. is the Lead and Copper Rule (LCR), which aims to reduce water corrosivity and limit leaching of lead or copper from potable water plumbing in buildings. The LCR sampling requires that the water sit at least 6 h in the premise plumbing before collection. However, most standard sampling protocols (e.g., the USEPA’s Total Coliform Rule for monitoring bacteria) require flushing for 3–5 min at taps within buildings prior to sample collection. The results reflect levels of bacteria in the water mains rather than in the building plumbing system.

1.2. Chloramine stability and reactions: bulk water, water mains, and premise plumbing

The formation of free ammonia when chloramine decays can cause a range of problems including nitrification, lower pH, increased availability of assimilable organic carbon, and enhanced regrowth (Edwards, 2005). Chloramine decay can occur due to reactions in bulk water, water mains, and building plumbing. The reactions tend to worsen with higher water age and other factors (Table 2).

1.2.1. Bulk water reactions

A range of bulk water characteristics can increase the rate of chloramine decay including increased concentrations of NOM, nitrite, or phosphate (Duirk et al., 2005; Margerum et al., 1994; Song et al., 1999; Valentine, 1985; Vikesland et al., 1998). Thomas (1987) showed that lowering pH by 0.7 units doubles the rate of chloramine decay. The chloramine decay rate has also been shown to double with each increase of 16 °C (Sathasivan et al., 2009). In contrast, increasing ammonia (NH_3) tends to increase the stability of chloramine in bulk solution (Margerum et al., 1994; Valentine, 1985).

1.2.2. Water mains reactions

Metals in the distribution system, such as copper, iron, and lead, can catalyze chloramine decay (Switzer et al., 2006; Vikesland and Valentine, 2002a; Zhang and Edwards, 2009) through reactions with pipe corrosion products (e.g., $\frac{1}{2}NH_2Cl + H^+ + Cu^{1+} \rightarrow Cu^{2+} + \frac{1}{2}NH_4^+ + Cl^-$) (AWWA, 2006). In at least some rare instances, chloramine has been shown to decay more rapidly than free chlorine in head-to-head tests with polyvinyl chloride (PVC), epoxy-lined copper, and stainless steel pipe materials (Edwards, 2005; Zhang and Edwards, 2009). Very high chloramine decay rates have also been noted to occur in some distribution systems (Edwards, 2005; Murphy et al., 1997; Powell, 2004).

Biofilm can grow on pipe surfaces and harbor microbes such as nitrifiers (Lipponen et al., 2004; Regan et al., 2007). Nitrification has been linked to rapid loss of chloramine compared to free chlorine in relatively inert pipe materials (e.g., PVC, epoxy-lined copper, new lead, stainless steel) (Zhang and Edwards, 2009). Maintaining a chloramine residual of at least 1–2 mg/L $NH_2Cl–Cl_2$ is thought to be sufficient to limit nitrifier growth in drinking water (Wolfe et al., 1990). Greater chloramine residuals may be required to inactivate bacteria in biofilms (Park and Kim, 2008).

1.2.3. Premise plumbing reactions

Premise plumbing can be especially susceptible to disinfectant decay compared to water mains because of the high pipe surface area-to-water volume ratios, dead ends, long detention times, higher temperatures, and lower disinfectant residuals (Table 1). One study found 60% decay of chloramine

Table 1 – Characteristics of U.S. public and private transmission systems (after Edwards et al., 2003; National Research Council, 2006).

Characteristic	Public infrastructure	Private infrastructure (premise plumbing)
Replacement value	\$0.6 trillion USD	>\$0.6 trillion USD
Pipe material	Cement, ductile iron, plastic, cast iron	Copper, plastics, galvanized iron, stainless steel, brass
Total pipe length (US)	0.97 million miles	>6 million miles
Approx. pipe surface per volume of water[b]	0.26 cm^2/mL[b]	2.1 cm^2/mL[b]
Complete stagnation	Relatively rare	Frequent
Disinfectant residual	Usually present	Often completely absent after stagnation
Flow	Relatively consistent	On/off
Temperature	0–30 °C	0–100 °C
Maximum cost over 30 yrs per consumer	\$500–7000 USD	Easily up to \$25,000 per homeowner or millions for buildings
Advocacy	Water industry (WIN)[a]	None

[a] Water infrastructure network.

[b] Assumed 15.2-cm diameter for mains and 1.9-cm diameter for home plumbing.

Table 2 – Conventional wisdom of chloramine decay and distribution system reactions.

Constituent/factor	Chloramine decay wisdom		
	Bulk water reactions	Water mains[a]	Premise plumbing[a]
Natural Organic Matter (NOM)	Accelerates decay	Reduces iron corrosion[b]	Reduces copper corrosion
Lower pH	Accelerates decay	Accelerates iron corrosion	Accelerates copper corrosion
Higher temperature	Accelerates decay	Increases metal pipe dissolution	Can increase or decrease metal release
Ammonia	Slows decay	No primary chemical effect; secondary effect of microbial regrowth and bacteria-induced corrosion	Stagnation often leads to nitrification and higher copper release[c]
Nitrite	Accelerates decay	Reduces iron corrosion[d]	Potential health concern[e]
Phosphate	Accelerates decay	Reduces iron and copper corrosion	Reduces copper corrosion

a Increasing iron and copper corrosion (i.e., amount of corrosion by-products) or dissolution would accelerate chloramine decay.
b (Elfström Broo et al., 1999).
c (Murphy et al., 1997; Zhang et al., 2009).
d (Pryor and Cohen, 1953).
e (Bruning-Fann and Kaneene, 1993).

in 6 h due to abiotic reactions with copper pipe (Zhang and Edwards, 2009), although the precise mechanism was not elucidated. Zhang et al. (2002) hypothesized that copper complexes with the ammonia in chloramine, increasing copper solubility and chloramine decay. However, in that work, chloramine decayed rapidly in high-density polyethylene (HDPE) bottles with and without copper. Others suggested that Cu^{2+} complexes with monochloramine to accelerate chloramine decay (Fu et al., 2009a, 2009b). A precise sequence of reactions was not proposed. Other investigators have noted that additional currently unidentified factors must be contributing (Krishna and Sathasivan, 2010).

1.2.4. *Chloramine and copper reactions*

Two hypothetical pathways are proposed to explain observations of abiotic chloramine disappearance. One reaction mechanism of high interest is based on an observation by Filimonov and Shcerbakov (2004). Filimanov hypothesized that Cu metal (Cu^0) in the presence of Cu^{2+} formed reactive cuprous ion (Cu^{1+} (Equation (1)). Cu^{1+} can subsequently react with nitrate (NO_3^-) to form nitrite (NO_2^-) and other reactive species (Equation (2)). Nitrite in the overall reaction (Equation (3)) could react further to decay chloramine (Equation (4)).

$$\frac{1}{2}Cu^0 + \frac{1}{2}Cu^{2+} \rightarrow Cu^{1+} \quad (1)$$

$$Cu^{1+} + NO_3^- + 2H^+ \rightarrow Cu^{2+} + NO_2^- + H_2O \quad (2)$$

$$\frac{1}{2}Cu^0 + \frac{1}{2}Cu^{2+} + NO_3^- + 2H^+ \rightarrow Cu^{2+} + NO_2^- + H_2O \quad (3)$$

$$NH_2Cl + NO_2^- + H_2O \rightarrow NH_4^+ + NO_3^- + Cl^- \quad (4)$$

In the second hypothetical pathway, Cu^{1+} forms and directly reacts with NH_2Cl (Equation (5)) to accelerate autodecomposition of NH_2Cl to ammonium ion (NH_4^+) and Cl^- (Bengough and May, 1924).

$$\frac{1}{2}Cu^0 + \frac{1}{2}Cu^{2+} \rightarrow Cu^{1+}$$

$$\frac{1}{2}NH_2Cl + H^+ + Cu^{1+} \rightarrow Cu^{2+} + \frac{1}{2}NH_4^+ + \frac{1}{2}Cl^- \quad (5)$$

$$\frac{1}{2}Cu^0 + \frac{1}{2}Cu^{2+} + \frac{1}{2}NH_2Cl + H^+ \rightarrow Cu^{2+} + \frac{1}{2}NH_4^+ + \frac{1}{2}Cl^- \quad (6)$$

These reaction pathways explain how the addition of Cu^{2+} (Fu et al., 2009a, 2009b; Nguyen and Edwards, 2005) could increase the rate of chloramine decay in the presence of metallic copper, especially during early stages of copper corrosion when low levels of Cu^{2+} are present in water. If both Cu^{2+} and NO_3^- in Equation (2) are present, the reactions (Equations (3) and (4), or Equation (6)) can cycle continuously until Cu^0 or NH_2Cl is depleted. Cu^+ and Cu^{2+} can both occur in potable water (Pourbaix, 1974).

This work is aimed at identifying and resolving issues of microbial regrowth resulting from rapid chloramine decay in new buildings with water conservation features. The contribution of new copper pipe to rapid chloramine decay and possible mitigation strategies are also examined.

2. Materials and methods

2.1. *Case study*

To identify and resolve drinking water taste and odor complaints in new buildings at UNC-CH, a systematic evaluation was conducted to determine the rate of chloramine loss, concentrations of microbes, and other related chemical parameters. Tests were carried out in two new buildings where complaints had occurred (sampling sites 1, 2, and 3) and a neighboring new building without taste complaints (sampling site 4). The building plumbing ranged from 5 to 11 months in age at the time of the first sampling event and when chemistry data were collected (Table 3).

After water in the test taps (sampling sites 1, 2, 3, and 4) was held stagnant for ≈16 h, 250 mL “first draw” samples were collected from each test tap for microbial analyses. To

Table 3 – Apparatus, experimental duration, and water quality information for each test conducted.

	Test			
	Case study[a]	pH, temperature, orthophosphate	Free chlorine vs. chloramine; copper pipe vs. glass; ammonia	Orthophosphate, Cu, Fe, Al, NO_3, TOC
Apparatus[b]	In-situ building plumbing on UNC campus[c]	30.5 cm length Cu pipe with 2.9 cm length solder[d]	46.0 cm length Cu pipe with 3.2 cm length solder[e]	
Test duration	48 h; building plumbing 5–11 months in age	57 d	33 d	127 d
Basic water condition	9 mg/L alkalinity as $CaCO_3$, 0.56 mg/L PO_4, 27 mg/L SO_4	34 mg/L alkalinity as $CaCO_3$, 17 mg/L Cl, 14 mg/L SO_4^{2-}, 2 mg/L Al, 0.3 mg/L TOC, and 1.25 mg/L NH_3–N	10 mg/L NO_3^- and 18 mg/L Cl	10 mg/L NO_3^- and 18 mg/L Cl
Additional parameters evaluated	Flushed line	0 and 3 mg/L P	0–4 mg/L NH_3–N (0.3 mM NH_3)	3 mg/L P, 2 mg/L Cu, 1 mg/L Fe, and 1 mg/L Al
pH	8.1	7 or 9.5	7	7
Temperature	Variable at tap	4, 20, or 40 °C	25 °C	25 °C
Disinfectants evaluated	0.1–3.9 mg/L NH_2Cl–Cl_2	4 mg/L NH_2Cl–Cl_2	4 mg/L Cl_2 or NH_2Cl–Cl_2 (0.06 mM Cl_2)	4 mg/L NH_2Cl–Cl_2 (0.07–0.14 mM Cl_2)
No. replicates	1	3	3	3
Measurements	Total chlorine, HPC, HABs, APBs, denitrifiers, *Legionella pneumophila*, *Pseudomonas aeruginosa*[f]	Total and free chlorine, metals (e.g., Pb, Cu)	Total and free chlorine, metals (e.g., Pb, Cu)	Total and free chlorine, metals (e.g., Pb, Cu)

a Typical water quality leaving water treatment plant.
b 1.9-cm diameter copper (Cu) pipe dipped in specified depth of molten 50:50 Pb–Sn solder.
c Cu pipe near faucet taps.
d 165 cm^2 Cu wetted surface area and 17 cm^2 50:50 Pb–Sn solder wetted surface area.
e 255 cm^2 Cu wetted surface area and 19 cm^2 50:50 Pb–Sn solder wetted surface area.
f HPC – heterotrophic plate count; HABs – heterotrophic aerobic bacteria; APB – acid producing bacteria.

quantify the rate of chloramine decay in building plumbing, extensive flushing of taps was conducted until water with a high chloramine residual (>2 mg/L as NH_2Cl–Cl_2) was drawn from the water main into each plumbing line tested. Thereafter, the water sat stagnant in the pipes. The chloramine residual was measured periodically by removing small aliquots from each tap approximately every hour for the first 8 h. A final sample was collected at 13–16 h. As a control and to test for bulk water reactions, tap water was withdrawn from test taps immediately after flushing and placed into a glass container. Chloramine residual was measured at similar time intervals as the test taps. In one sampling event, HPC samples were collected before and after tap flushing throughout the same building as sampling taps 1 and 2.

2.2. Bench-scale tests

Complementary bench-scale testing was conducted to document the mechanisms of chloramine decay in water contacting copper plumbing.

2.2.1. Experimental apparatus procedure

New pipes were used in bench-scale tests to eliminate other factors associated with harvested pipes such as 1) biofilms with differing microbial populations on the pipe surface, 2) trace surface catalysts and rust deposits invariably present on old pipe, 3) flux and other installation debris on building plumbing, and 4) other uncertainties of prior exposure of harvested pipe. Silicone stoppers were used to seal the pipes, and bench-scale tests were conducted in triplicate.

In one set of experiments evaluating the effects of NH_3, Cu^{2+}, NO_3^-, Al^{3+}, Fe^{3+}, and NOM on chloramine decay, a 46-cm length of copper pipe (1.9 cm ∅) with a 3.2 cm length of solder coating was used. In a second set of tests a 30.5-cm length copper pipe (1.9 cm ∅) with a 2.9 cm length of solder was used to investigate effects of pH (7 or 9.5), temperature (4, 20, or 40 °C), and orthophosphate addition on the rate of chloramine decay. The solder was present to evaluate a related problem with elevated lead in drinking water (Elfland et al., 2010), and preliminary testing indicated its presence had very little impact on the rate of chloramine decay. To examine trends as the copper pipes aged, the experiments were run for 33, 57, or 127 days (Table 3). Water in all pipes was changed Monday, Wednesday, and Friday each week. To compare chloramine decay in the presence of an inert surface versus copper pipe, tubes made of pure glass were tested in parallel for a subset of water conditions: (1) NH_2Cl, (2) NH_2Cl and Cu^{2+}, and (3) NH_2Cl, Cu^{2+}, and P. The test water also contained 10 mg/L NO_3–N and 18 mg/L Cl (Table 3).

2.2.2. Water chemistry

In all bench-scale experiments with soldered pipes, test waters were prepared using reagent grade salts added to distilled and deionized water (Table 3). Target concentrations (Table 3) of free chlorine or chloramine (4:1 mg Cl_2 to mg N ratio) were prepared in the respective test waters by adding appropriate volumes of 6% (by weight) sodium hypochlorite (NaOCl) and 14.8 N ammonium hydroxide (NH_4OH) to form monochloramine. The pH was then adjusted to target values (Table 3) using hydrochloric acid (HCl) or sodium hydroxide (NaOH). Soldered pipes were tested in triplicate using the prepared water.

In the experiments investigating the effects of pH and temperature, the following additional adjustments were made to the test water: alkalinity of 34 mg/L as calcium carbonate ($CaCO_3$; 0.68 mM $NaHCO_3$), 0.24 mM calcium chloride ($CaCl_2 \cdot 2H_2O$), 0.15 mM calcium sulfate ($Ca_2SO_4 \cdot 2H_2O$), 0.07 mM pre-formed aluminum solids as Al ($Al_2(SO_4)_3 \cdot 18H_2O$, 2 mg/L Al), 0.3 mg/L total organic carbon (TOC) from prechlorinated NOM, and 0.089 mM ammonia (1.25 mg/L NH_3–N) (Table 3). The chlorine demand of the NOM (rinsed Aldrich humic) was met by dosing 0.43 mg/L Cl_2 (6 μM) per mg/L TOC before use in experiments, which decayed to 0 mg/L Cl_2 24 h later. Orthophosphate (0 or 3 mg/L as P; Na_2HPO_4) was varied in the test water (Table 3). Pre-formed aluminum solids were prepared in accordance with other work (Rushing and Edwards, 2004).

To evaluate the effect of ammonia concentrations on chloramine decay, ammonia varying from 0 to 4 mg/L N (0–0.3 mM NH_3) was added to water containing 0.16 mM sodium nitrate ($NaNO_3$), 0.50 mM sodium chloride (NaCl), and 4 mg/L total chlorine as NH_2Cl–Cl_2 (0.06 mM NH_2Cl–Cl_2), and tested at pH 7.0 and 25 °C (Table 3). In addition, either 0.097 mM PO_4 (Na_2HPO_4, 3 mg/L P), 0.03 mM cupric chloride ($CuCl_2$, 2 mg/L Cu), 0.018 mM Fe (1 mg/L Fe), or 0.037 mM aluminum solids (1 mg/L Al) was added as discussed in later results to determine their impact on disinfectant decay.

2.3. *Analytical methods*

Free and total chlorine were measured on a Hach DR 2700 spectrophotometer according to Standard Method 4500-Cl (APHA, 1998). The pH of the bulk water was measured with an Accumet electrode in accordance with Standard Method 4500-H^+ B (APHA, 1998). Unfiltered samples from the bench-scale tests were measured for copper and lead using inductively coupled plasma emission spectroscopy (ICP-ES).

Bacterial colonies including heterotrophic aerobic bacteria (HAB), denitrifying bacteria, and acid producing bacteria (APB) were tested using standardized colorimetric test kits (Cullimore, 2010). Heterotrophic plate counts (HPC) were determined using the spread plate method with R2A agar or the SimPlate method (APHA, 1998). The two HPC methods can produce different values; therefore, comparisons of trends are made only within datasets produced using the same method. HABs and HPCs are useful indicators of microbial activity. To analyze for *Legionella pneumophila*, collected samples were acid pretreated (Strickhouser, 2008) and cultured on buffered charcoal yeast extractor (BCYE) agar. *Pseudomonas aeruginosa* was cultured on cetrimide agar, incubated at 37 °C for 24 h, and counted (Strickhouser, 2008).

Statistical analysis including paired t-testing, linear regression, and ANOVA were used to determine the significance of the findings, with appropriate corrections using Bonferroni's method (Bonferroni, 1935).

3. Results and discussion

3.1. *Initial identification of a problem*

After taste and odor complaints by occupants in two UNC-CH buildings with water conservation features, testing revealed no detectable chloramine residual (<0.17 mg/L NH_2Cl–Cl_2) in first draw samples from some taps. Extensive flushing was then implemented at the sampling sites until water with a high chloramine residual (>2 mg/L NH_2Cl–Cl_2) was drawn from the water main into each line. Some distal taps in the buildings (e.g., sampling site 4) had to be flushed 40 min before the disinfectant levels approached those present in the water main (Fig. 1a). At sampling sites 1 and 2, the taps were flushed 15 min before disinfectant levels were above 3 mg/L NH_2Cl–Cl_2 (Fig. 1a). The maximum disinfectant level at sampling site 3 (2.5 mg/L NH_2Cl–Cl_2) was reached after 5 min of flushing.

To measure the rate of chloramine decay, water was allowed to stagnate in the building plumbing after flushing. Aliquots were withdrawn periodically, and the chloramine residual was measured. At each sampling site, 78–91% of the disinfectant disappeared in 6 h and completely disappeared within 12–18 h (Fig. 2). To rule out bulk water reactions exerting a chloramine demand, water was collected immediately after flushing and allowed to stagnate in a glass

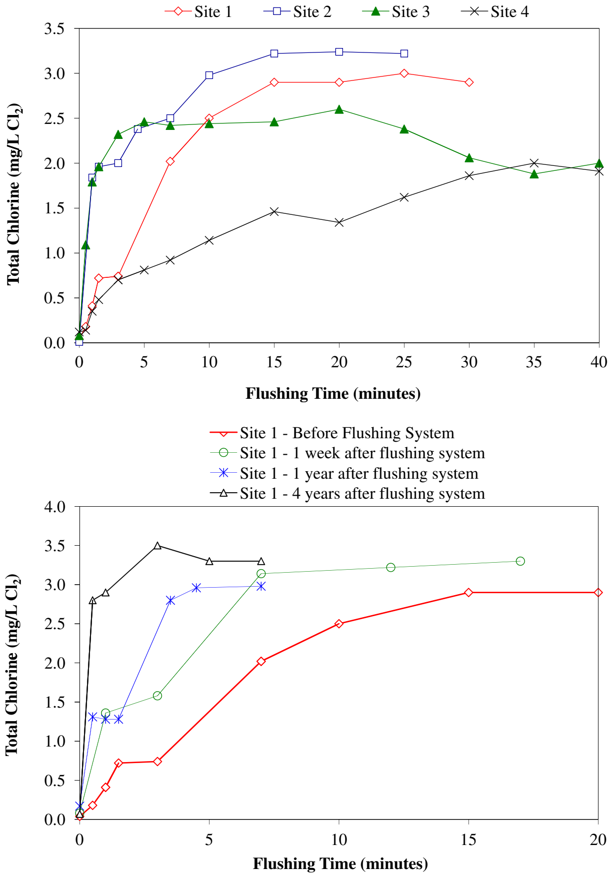

Fig. 1 – Chloramine concentration during flushing for (A) four sites and (B) sampling site 1 before and after installation of flushing system.

container. Little disinfectant decay occurred (Fig. 2). These observations counter the widely held view, based on testing in water mains, that chloramine residual is typically very stable and persistent. Instead, the findings support the hypothesis of Zhang et al. (2009) that rapid chloramine decay can occur in some building plumbing.

The lack of chloramine residual was suspected to be due to abiotic factors because there was no evidence of nitrification (pH increased during stagnation; little or no nitrite and nitrate were produced). It is more likely that chloramine was decaying in this water via Equation (6), which does not require the presence of nitrate. The calculated water residence times in the building plumbing exceeded several days even during full occupancy, which is attributed to the design of the plumbing systems and the reduced potable water demand. The flushing test indicated that the distances from sites 1 and 2 to the main were comparable. However, chloramine decay was more rapid at site 1 (Fig. 2), where the plumbing system riser lacked toilets. This lack of heavy water demand resulted in higher water age compared to site 2 (where toilets are present, were in-use, and could pull fresh water into the line). These factors at site 2 marginally improved the persistence of chloramine, but 95% of the residual still decayed in 16 h.

Relative to a comparable older building, potable water consumption was 60% less in the newer building that incorporated green plumbing features (sites 1 and 2) (Table 4). Similarly, the sampling site 4 building that uses both green plumbing devices and non-potable use for toilets had a 90% reduction in potable water use compared to comparable older buildings. The 90% reduction in water use translated to a 10 times increase in water age, given that design diameters of premise plumbing pipes were not reduced for the buildings with water conservation features. It was also noted that the water in buildings with water conservation features tended to have higher temperatures (up to 30 °C), due to longer stagnation times and water fountain chillers being installed at greater distances from the bubblers in water fountains associated with the taste and odor complaints. It has also been established that new copper piping can accelerate chloramine decay rates (Nguyen and Edwards, 2005; Zhang and Edwards, 2009).

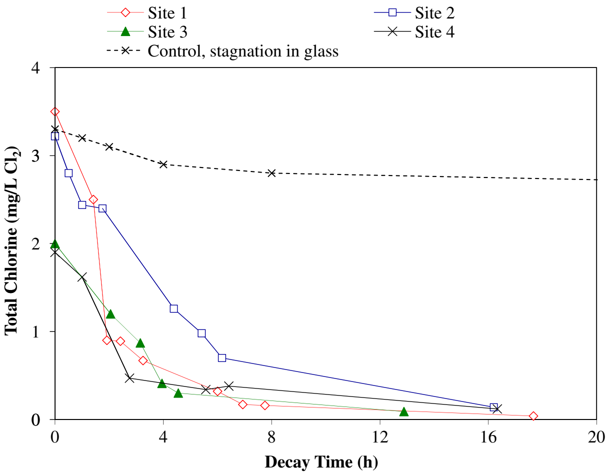

Fig. 2 – Chloramine decay for four sampling sites.

3.2. *Characterization of bacteria levels*

Testing of heterotrophic plate count (HPC) bacteria and other microbes was conducted, due to the presence of low disinfectant residuals that could allow microbial regrowth (Rittman and Snoeyink, 1984; Sathasivan and Ohgaki, 1999). Testing of other taps in the same building as sampling sites 1 and 2 revealed very high levels (>308,000 cfu/mL) of HPC bacteria in several first draw water samples, which is not unexpected given the warm conditions, lack of chloramine residuals, and long residence times. These levels are orders of magnitude greater than the threshold concentration of 500 cfu/mL often used to indicate excessive microbial regrowth in water mains (USEPA, 1989). A significant negative correlation (df = 46, p-value<0.01) was found between the logarithm of HPC and the chloramine concentration (Fig. 3). That is, when the chloramine concentration was less than 1 mg/L $NH_2Cl–Cl_2$ after stagnation, the HPC concentration was typically 560–140,000 cfu/mL. Following a 1-min flush at the sampling taps, the chloramine concentration increased about 0.7 mg/L $NH_2Cl–Cl_2$, and the water contained 60–99.9% fewer HPCs (up to 3-log reduction) for 20 out of 24 samples (Fig. 3).

More detailed bacterial analysis also was conducted for the four sampling sites. Human pathogens including *P. aeruginosa* and *Legionella* were not detected in any of the samples despite the absence of disinfectant. However, concentrations of HPCs, heterotrophic aerobic bacteria (HAB), denitrifying bacteria, and acid producing bacteria (APB) were sometimes elevated. Greater than 87,000 cfu/mL of HPC bacteria were observed at the sampling sites after overnight stagnation. This concentration of HPC does not violate any health standards but is of concern due to the potential influence on taste and odor. Denitrifiers, which can convert nitrate to nitrite in water, were as high as 200,000 cfu/mL at sampling sites 1 and 4. Over 500,000 cfu/mL of APBs was measured at sampling site 1, which was of interest due to high levels of lead release from brass plumbing devices observed in this building a few months prior to this study (Elfland et al., 2010). Although microbial regrowth may also occur for sites using free chlorine disinfectant, the impact may be less significant because free ammonia nutrients are not formed after chlorine decay. Given all of the negative consequences associated with loss of chloramine residual, remedial actions were implemented.

3.3. *Remediation*

An automated flushing system was installed near sampling site 1 to bring fresh water into the plumbing every 6 h. While automated flushing seems incompatible with water conservation, a relatively small amount of flushing was needed to markedly increase disinfectant residuals (flushing represented less than 1% of the daily water use in the building). In other situations, manual flushing was triggered by consumer taste and odor complaints, and water fountains were replaced with models having integrated chillers to reduce water temperatures.

The automated flushing increased the disinfectant residual (closer to 2 mg/L) in pipes. Only 3 min of flushing at the sampling site 1 tap was required to achieve 2 mg/L $NH_2Cl–Cl_2$ compared to 7 min before automated flushing was installed

Table 4 – Comparison of water use in green and conventional buildings.

Building type	Description	Average potable water use/area (gal/ft^2/month)	Green plumbing devices
Newer lab building (comprises Sites 1 and 2)[a]	Green plumbing devices	26	Yes
Older lab building[b]	Conventional	63	No
Newer classroom building (Site 4)	Green plumbing devices, all potable water use	7	Yes
	Green plumbing devices, non-potable water for toilet flushing	1	Yes
Older classroom building[b]	Conventional	10	No

a Site 1 - Riser with no toilets, Site 2 - Riser with toilets.
b Building used as baseline for water use comparison.

(Fig. 1b). Microbial levels also declined sharply after flushing was implemented. HABs decreased 1.4-log immediately after the flushing system installation and were undetectable after 4 years (Fig. 4). APB dropped about 2-log from 505,000 cfu/mL to 5000 cfu/mL and also were undetectable after one year. Denitrifiers were less impacted by the more persistent chloramine residual; the detected population declined less than 1-log over 4 years (Fig. 4).

HPC and other standard microbial measurements are useful indicators of aesthetic concerns and can be indicators of potential health risks arising from deficiencies in plumbing system design (Borella et al., 2004; CDC, 2008). Fortunately, although HPCs were high in some of the buildings studied, opportunistic pathogens such as *Legionella* or *P. aeruginosa* were not detected in water from the building plumbing systems.

While no adverse health consequences have been documented thus far, prudence and aesthetic concerns would suggest that steps should be taken to mitigate microbial regrowth. First and foremost, more research is needed on premise plumbing system designs and remedial measures to prevent deterioration of water quality. Reducing water residence times in buildings via design and operational changes is one avenue. Alternatively, it is possible that implementing nutrient-limitation strategies at water treatment plants (i.e., limiting ammonia and organic carbon) could be effective, but very little research on this topic has been conducted for premise plumbing. Finally, process engineering tools such as computer models to determine water residence time and loss of disinfectant residual in distribution systems (e.g., Rossman, 2000), might be adapted to help operate and maintain premise plumbing systems.

In the buildings studied, remedial strategies including installation of an automated flushing system helped maintain a chloramine residual in the plumbing. Microbial regrowth dramatically decreased, and taste and odor complaints were also resolved. The rate of chloramine decay in the plumbing remained rapid even one year after the initial test but slowed by 50% four years after implementing the flushing system near site 1. Overall, the field studies indicated that a combination of new copper piping and high water age contributed to problems observed in the new buildings, instigating additional tests at bench-scale to examine abiotic paths of chloramine decay.

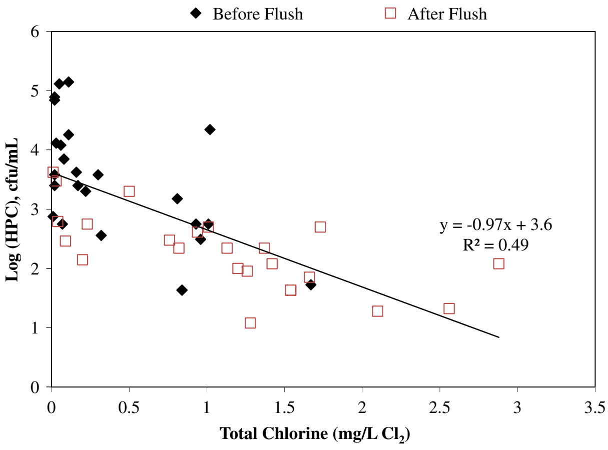

Fig. 3 – Heterotrophic plate count (HPC) and total chlorine concentration before and after flushing in one building. The HPC samples were collected after periods of stagnation (i.e., before flushing at the tap) at various taps and after 1 min of flushing at each tested tap. The Simplate method was used for HPC analysis.

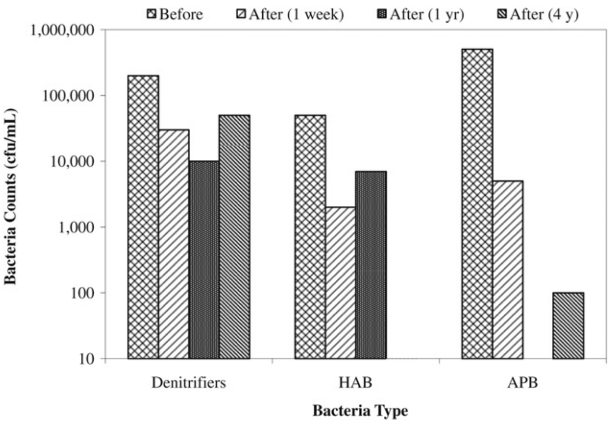

Fig. 4 – Testing of denitrifiers, heterotrophic aerobic bacteria (HAB), and acid producing bacteria (APB) before and after implementation of regular flushing (every 6 h) in sampling site 1 with microbial regrowth problems.

3.4. Comparison of free chlorine and chloramine decay in glass and copper pipe

In inert glass containers, free chlorine and chloramine disinfectants had very little decay over 8 h (Fig. 5), consistent with prior results for stable disinfectants in bulk water or water mains. The test water contained 10 mg/L NO_3–N and 18 mg/L Cl as well as free chlorine or chloramine (4:1 mg Cl_2/mg N) disinfectant (Table 3). However, both free chlorine and chloramine in pH 7 water decayed rapidly over time in copper pipe (Fig. 5). At least 85% of the initial chloramine residual (4 mg/L NH_2Cl–Cl_2) decayed within 8 h, whereas 100% of the free chlorine residual decayed in 8 h. Moreover, addition of greater concentrations of ammonia, which has been shown to stabilize chloramine residuals in bulk water (Margerum et al., 1994), had little impact on the rate of chloramine decay in copper pipe (Fig. 5).

The observed initial rate of chloramine decay was between 4.5×10^4 and 7.0×10^4 $(M\cdot h)^{-1}$. These rates are more than two orders of magnitude (136 times) greater than the fastest decay measured by Fu et al. in pH 6.1 water with 1 mg/L Cu^{2+} (Fu et al., 2009b). The chloramine decay reaction occurring in this work was best described as first order decay, which was approximately 0.29 h^{-1} for chloraminated water contacting copper pipe up to 33 days of age. Hence, the reaction of chloramine with copper pipe best explains the rapid chloramine decay observed in the new buildings (Fig. 2). Although others suggested that ammonia can complex with copper to increase copper solubility and chloramine decay (Zhang et al., 2002), in this work there was no observable difference in chloramine decay with increasing ammonia concentrations (Fig. 5). Free chlorine decay was more than two times faster (0.66 h^{-1}) than chloramine decay in copper pipe.

3.5. Effects of pH, temperature, and phosphate on chloramine decay

Chloramine decayed more rapidly at pH 7 than at pH 9.5 in copper pipe. The chloramine residual, initially 3.8 mg/L NH_2Cl–Cl_2, depleted 66–79% in 1 h at pH 7, whereas chloramine decayed no more than 10% after 1 h of stagnation in pH 9.5 water (Fig. 6). Copper tends to corrode faster with decreasing pH (Schock et al., 1995), which can subsequently exert a chloramine demand as suggested in Equation (6).

Higher temperatures also markedly increased the rate of chloramine decay. At pH 9.5 and 40 °C, the chloramine residual decreased from 3.8 mg/L NH_2Cl–Cl_2 to 1 mg/L NH_2Cl–Cl_2 (i.e., 74% decrease) in 4 h (Fig. 6). In contrast, 29% of the residual decreased for the same pH at either 20 °C or 4 °C after the same time period. Although chloramine residual typically becomes more stable at lower temperatures, this result suggests that copper catalysis of chloramine decay can occur at relatively low temperature (4 °C). At pH 7, temperature played a lesser role, as chloramine decayed 93–97% after 4 h at pH 7 and temperature ranging from 4 to 40 °C (Fig. 6).

Orthophosphate is thought to decrease chloramine stability in the bulk water (Margerum et al., 1994). However, the addition of the orthophosphate corrosion inhibitor (3 mg/L P) significantly reduced the rate of chloramine decay in copper pipe (Fig. 7). Specifically, dosing orthophosphate slowed the chloramine decay rate 82–99.5% from 6.4×10^4 $(M\cdot h)^{-1}$ to 1.2×10^4 $(M\cdot h)^{-1}$ and 1.7×10^5 $(M\cdot h)^{-1}$ to 7.8×10^2 $(M\cdot h)^{-1}$ for

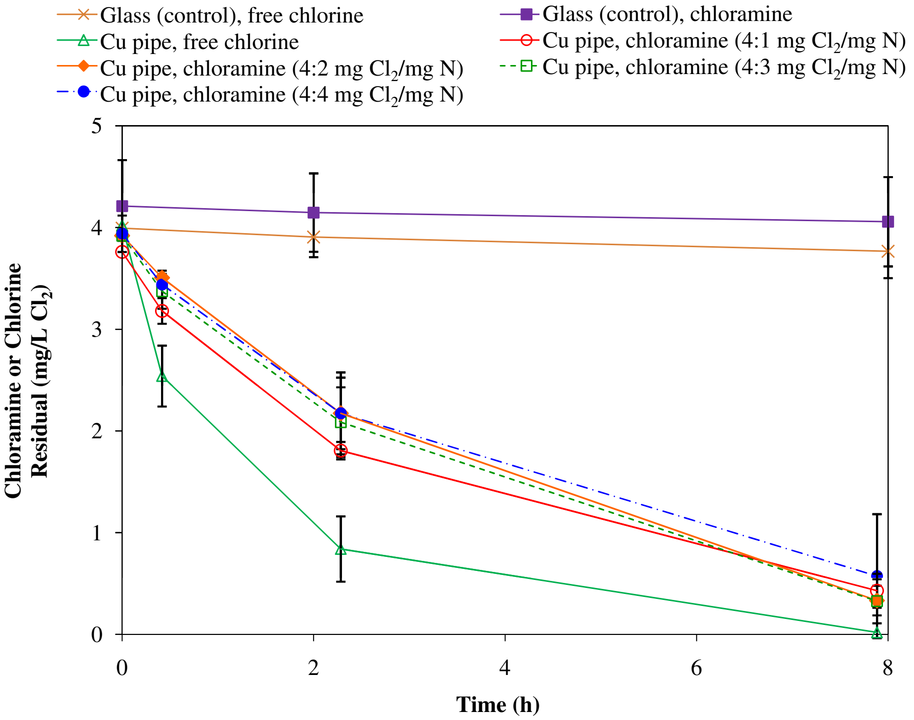

Fig. 5 – Chloramine or free chlorine residual for 0–4 mg/L NH_3–N as a function of stagnation time in glass containers or copper pipes. The test water was at pH 7 and 25 °C. Data points are average of triplicates during day 12 of the study. Similar trends were observed at day 33. Error bars represent 95% confidence intervals.

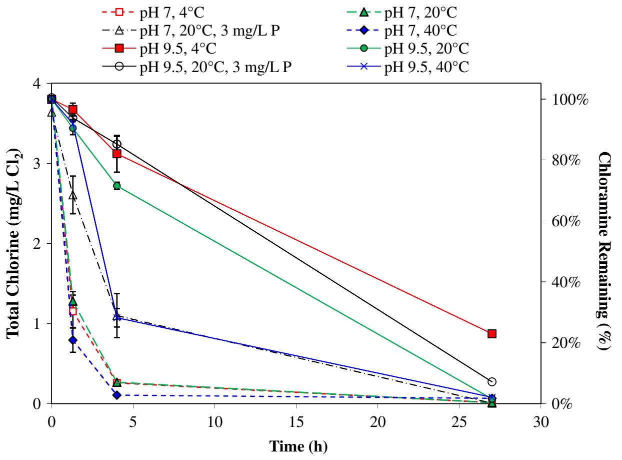

Fig. 6 – Chloramine residual remaining in potable water after 0–27 h of stagnation in copper pipe. Orthophosphate (0 or 3 mg/L P), pH (7 or 9.5), and temperature (4 °C, 20 °C, or 40 °C) were tested. Data points are average of triplicates during week 2. Error bars represent 95% confidence intervals.

pH 7 and 9.5, respectively. Orthophosphate presumably increased the stability of chloramine (Fig. 7) by lessening copper corrosion and the mass of copper released to water (results not shown). This would also decrease the amount of copper reacting via Equation (3) or (6).

3.6. *Effects of cupric, nitrate, aluminum, and iron*

After establishing the effects of temperature, pH and phosphate, more lengthy tests (127 d) were used to examine chloramine decay in copper pipe in the presence of Cu^{2+}, Al^{3+}, NO_3^-, and Fe^{3+}. These species have been found to influence disinfectant residual stability (Fu et al., 2009b; Ives and Rawson, 1962; Vikesland and Valentine, 2002a, b). Nitrate and iron (1 mg/L Fe) had no significant effect on chloramine decay in copper pipes.

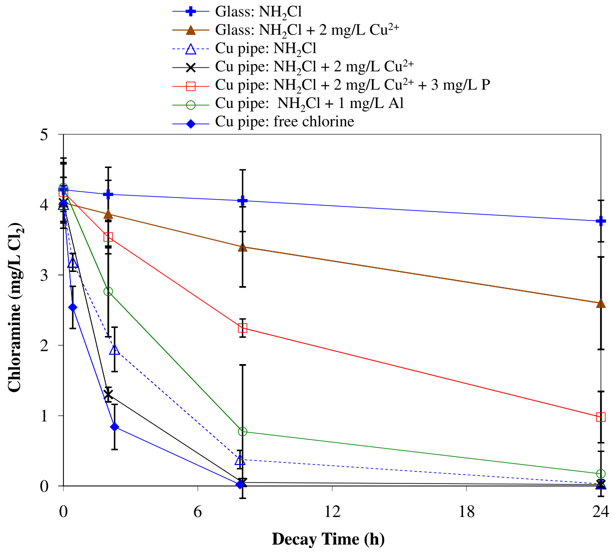

Fig. 7 – Chloramine concentration remaining during 24 h of stagnation in glass or copper pipes. Data are average values from decay curves conducted at day 70, 82, and 127. The test water contained 10 mg/L NO_3^- and 18 mg/L Cl at pH 7. Error bars represent 95% confidence intervals.

When 2 mg/L soluble Cu^{2+} was added to chloraminated water contacting glass, the chloramine decay rate increased four times (0.02 h^{-1} compared 0.005 h^{-1} without Cu^{2+}) (Fig. 7). However, the chloramine decay rate increased 60 times (0.29 h^{-1} vs. 0.005 h^{-1}) when contacting copper pipe with no initial Cu^{2+} dosed. In the presence of the copper pipe, addition of 2 mg/L Cu^{2+} accelerated the rate of chloramine decay by 70% compared to water with no initial Cu^{2+} (Fig. 7). The chloramine decay rate in copper was orders of magnitude greater than in glass (0.005 h^{-1} for glass vs. 0.346 h^{-1} for copper pipe). Clearly, while the addition of Cu^{2+} accelerated chloramine decay (Fig. 7; Fu et al., 2009b), the copper pipe corrosion (and possibly released Cu^{1+}) greatly accelerated chloramine decay. Similar to findings in the shorter test (Fig. 6), dosing orthophosphate corrosion inhibitor (3 mg/L P) decreased the chloramine decay rate by seven times in copper pipe (Fig. 7). Specifically, the chloramine residual decreased 89% after 8 h in pH 7 water initially containing 4 mg/L $NH_2Cl{-}Cl_2$ and 2 mg/L Cu^{2+} contacting copper pipe. Only 45% of the chloramine residual decayed in the same conditions with the addition of 3 mg/L P.

Although aluminum had little effect on chloramine decay after 70 d of aging, copper pipes exposed to 1 mg/L Al had drastically slower chloramine decay at 127 d (0.09 h^{-1} vs. 0.30 h^{-1}). This is presumably because aluminum gradually coated the copper surface to form a barrier between the copper and chloramine. In contrast, decay rates for the other test conditions did not change significantly throughout the test (results not shown).

The rapid loss of chloramine residual in some waters that contact newer copper pipe is of concern from the perspective of aesthetics (taste and odor) and possibly health. Older pipes, thicker scales, presence of silica, and presence of bicarbonate can reduce the extent of this problem. Factors that could negatively affect copper passivation include lower water use in buildings, high bacterial populations, and use of corrosive flux in installation. In practice, copper pipe often forms a passive film, which inhibits chloramine decay. The amount of time required for passivation of copper varies widely (can occur within minutes or may never occur during the lifetime of the pipe). Related work also demonstrates reduced rates of free chlorine decay in the presence of copper pipe in certain water conditions (Nguyen et al., 2011). In that work, free chlorine decay in low alkalinity water contacting copper pipe could be retarded within one week after orthophosphate was dosed or after the alkalinity or pH was increased. These reactions are deserving of future practical research.

4. Conclusions

- Rapid chloramine decay and microbial regrowth were observed in new buildings incorporating advanced water conservation features. More than 40 min of flushing was required at some taps to achieve chloramine levels measured in the water mains. When contacting copper pipes, the chloramine disinfectant decayed as much 90% in 6 h and completely disappeared after 12 h.

- Less microbial regrowth (2-log) occurred after implementation of an automated flushing system bringing a relatively small volume of fresh water into the plumbing line, which in turn helped maintain higher chloramine residuals. Although chloramine decay was relatively rapid one year after the initial testing, the rate had slowed by 50% after 4 years. It is possible that relatively low water flow through the plumbing delays the natural aging process.
- Bench-scale testing revealed that copper pipe catalyzed rapid chloramine decay. Dosing Cu^{2+} to the water slightly accelerated chloramine decay, but the greatest impact was from copper pipe corrosion.
- From the perspective of chloramine decay in bulk water, greater ammonia concentrations and lower phosphate concentrations reduce chloramine decay. Phosphate corrosion inhibitor interferes with reactions between chloramine and copper pipe. This results in substantial overall benefits from phosphate in maintaining chloramine residuals in premise plumbing. Aluminum deposits could also maintain chloramine residuals after long exposure times. It is possible that different trends would be observed in different waters.
- Lower pH and higher temperature accelerated the chloramine decay reaction in copper pipe.
- Design of water systems in buildings should consider impacts of lower water use (higher water age) on microbial regrowth, and consider reducing pipe diameter or ensuring minimal flushing to maintain palatable water.

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Appendix. Supplementary data

Supplementary data related to this article can be found online at doi:10.1016/j.watres.2011.11.006.

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