Factors Contributing to Chlorine Decay and Microbial Presence in Drinking Water Following Stagnation in Premise Plumbing

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Abstract

Premise plumbing is the part of the drinking water distribution system closest to the point of use. Since premise plumbing is characterized by a long residence time, elevated temperature, and reduced levels of disinfectant residue, drinking water in premise plumbing typically experiences elevated levels of microbial presence as compared to finished water exiting water treatment utilities, particularly under stagnation conditions frequently encountered in premise plumbing. Thus, stagnant drinking water in premise plumbing may represent an important source of public health risk. Therefore, the objective of this study is to identify factors contributing to the deterioration of microbiological quality of stagnant drinking water in premise plumbing. Results from this study indicated that the service age of premise plumbing system is positively correlated to the concentration of microorganisms in stagnant drinking water; Another factor contributing to microbial contamination is the usage pattern, with systems experiencing lower levels of water consumption exhibiting greater microbial contamination than those having greater water usage patterns; Since disinfectant residue is an important determinant of microbial contamination, the loss of free chlorine as the most common disinfectant residue was further examined. My results demonstrate that pipe material has significant impact on the decay rate of free chlorine, with copper pipe showing the greatest chlorine decay rate, and PVC pipe showing the slowest. The deposits onto the pipe wall appear to reduce the rate of chlorine decay, likely forming a barrier

between the pipe material and water, which slows down the reaction between the pipe wall and the disinfectant. Moreover, pipe diameter and temperature could significantly influence the rate of chlorine decay, with greater diameter leading to smaller surface-to-volume ratio and subsequently a slower chlorine decay rate. As expected, elevated temperature was shown to accelerate chlorine loss. These results provide important insights into the mechanisms of chlorine decay in premise plumbing and the factors contributing to the deterioration of the microbiological quality of drinking water in premise plumbing, which could facilitate the development of effective strategies for controlling water quality in premise plumbing and reducing public health risks from waterborne infectious diseases.

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List of Acronyms and Abbreviations

ANOVA	analysis of variance
C	chlorine concentration at time t (mg/L)
Cfu	colony-forming unit
Co	initial concentration
CPVC	chlorinated polyvinyl chloride
DI water	distilled and deionized water
DPD	N,N-diethyl-p-phenylenediamine
EPA	Environmental Protection Agency
HPC	heterotrophic plate count
MDPE	medium density polyethylene
PEX	cross-linked polyethylene
PVC	polyvinyl chloride
R^2	coefficient of determination
k	total decay constant (hr^{-1})
k_b	bulk water chlorine decay constant (hr^{-1})
k_w	pipe wall chlorine decay constant (hr^{-1})

t	time
WHO	World Health Organization

INTRODUCTION

Nationwide, 34 billion gallons of drinking water are treated everyday in the United States and around 63% is by consumers for end use such as consumption, cleaning and irrigation *(NRC, 2006).* Drinking water treatment plants remove or reduce contaminant levels to meet drinking water quality standards for the intended use. Treatment processes include physical settling, filtration, chemical coagulation and disinfection. Suspended solids, microbes, algae, fungi, minerals, fertilizers, and metallic ions are the primary contaminants removed in water treatment *(Water purification-Wikipedia)*. Drinking water treatment plants provide treated water to the consumer's tap by the drinking water distribution system, which includes a wide variety of pipes, pumps, valves, reservoirs, and other hydraulic components. There are lots of potential issues, such as backflow, contamination during installation, maintenance and operation. Thus, maintaining the desired water quality during the distribution and transmission processes is necessary and essential for human health. Engineers define premise plumbing as the pipe portion between service connection line and final pipes of end-use *(Kelly A. Reynolds, 2008).* As a part of the drinking water distribution system, premise plumbing deserves more attention due to its longer water residence times, more frequent stagnation conditions, and elevated aqueous temperature *(NRC, 2006).*

Researchers determined several unique characteristics of premise plumbing compared to those of the main distribution systems *(NRC, 2006)*:

- Larger surface-to-volume ratio,
- More pipe materials utilized,
- Better reservoir for bacterial and potential pathogens growth,
- Greater water age,
- Greater accumulation of deposits,
- Elevated temperature,
- Disinfectants loss.

Although drinking water treatment plants do not have direct responsibility for the changes in water quality of premise plumbing, customers' negative feedback could affect the performance of treatment plants *(Traci Case, 2009).*

Drinking water is delivered through distribution and transmission systems and then enters premise plumbing. Based on a variety of chemical, physical and microbial mechanisms, contaminants accumulate and are released in water, especially in that of premise plumbing when water flow stagnates. Because of bacterial re-growth in the faucet aerator of premise plumbing, (*Lechevallier, 1980*) bacterial density in stagnant water is two to three orders of magnitude greater than that in flushing water *(Brazos et al., 1985).* However, little work has been specifically focused on the high density of heterotrophic bacteria in premise plumbing systems, representing an important knowledge gap for the control of waterborne diseases from drinking water consumption. Biofilm always exist between the bulk water and pipe wall where microbes could grow. For this reason, biofilm may further contribute to the deterioration of the microbiological quality of drinking water. Previous research has revealed that 95% of the microbes are present on the pipe wall as biofilm, while the remaining 5% are present in bulk water *(Flemming et al., 2002)*. Some microbes with pathogenic characteristics may also exist in biofilm *(Percival et al., 2000)*. For example, in the biofilms of premise plumbing systems, *Legionella spp.* and nontuberculous *Mycobacterium spp.* have been detected and thus are considered to be potential pathogens *(Tobin-D'Angleo et al., 2004; Flannery et al., 2006; Tsitko et al., 2006)*. Therefore, stagnant water is an ecological niche for various microbes and thus fosters biofilm production *(Snoeyink et al., 2006)*. Previous research stated the bacterial re-growth process as following *(Caroline Nguyen et al., 2008)*:

Organic Carbon + Nitrogen + Phosphate + Trace Nutrients + O_2 → Heterotrophic bacteria re-growth

Disinfectants are added in treated water to control heterotrophic bacterial re-growth in premise plumbing. Most drinking water treatment plants choose free chlorine and chloramine as the main disinfectants to protect water quality in treated drinking water. However, free chlorine has been used more widely than chloramine due to its stronger oxidizing property and a variety of benefits such as microorganism inactivation, taste and odor control, and metallic ions removal. Based on the following chemical reactions, added chlorine forms hypochlorous (HOCl) and hydrochloric (HCl) acid and consequently dissociates into H^+ and OCl^-.

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^-$$

$$HOCl \rightarrow H^+ + OCl^-$$

Chlorine concentration in treated water is not constant in premise plumbing and decreases as water residence time increases *(Clark et al., 1994)*. The direct result of disinfectant loss is to increase and accumulate microorganisms in the water or on the pipe wall.

A large range service ages of premise plumbing systems constructed in the United States, even the pipes from the late 19th century, are still used at present *(NRC, 2006)*. As pipes age, premise plumbing becomes susceptible to corrosion accumulation, biofilm development, and even changes in pipe physical and chemical characteristics. Service age of premise plumbing could affect water quality *(Kelly A. Reynolds et al., 2008)*. As the age in pipe service increases, the rate of disinfectant loss increases *(Patrick Asamoah Sakyi1, 2012)*, resulting in premature water stagnation. Thus higher bacterial density could appear if the water was stagnant in premise plumbing *(Maul et al. 1985; J. Wingender, 2004)*. Moreover, as pipes age, undesired deposits could accumulate in bulk water or on the pipe wall. Recent research determines that the primary deposits in drinking water premise plumbing include minerals, organic matters, corrosion products, and biofilm *(Vikas Chawla, 2012).* These deposits have adverse effects not only on flow of water transmission *(S.A. Imran, 2005)* but also on water quality, because deposits may consume free chlorine *(B. Kowalska et al., 2006; Zhang et al., 1992; DiGiano and Zhang, 2005).* Usage of premise plumbing, defined as flushing frequency, is also critical for water quality. Flushing strategy, known as usage pattern, could effectively wash away undesired contaminants to keep the water clean *(Z. Michael Lahlou, 2002)*. Additionally, 95% of the potential microbial contaminations were from biofilm on the inside surface of the pipe: higher usage patterns could flush microorganisms and reduce biofilm accumulation, destroying habitats for bacterial re-growth *(J. Wingender, 2004)*.

The survey of *AWWA (2002)* indicates that premise plumbing mainly uses polyethylene, galvanized steel, PVC and copper pipes for different purposes. Older premise plumbing uses lead service joints and brass pipes with high lead concentration, while newer systems often use raw materials *(Traci Case, 2009)*. Although around 90% of drinking water premise plumbing use copper pipes *(Juneseok Lee, 2008)*, the pitting corrosion of copper pipes is considered expensive to maintain and repair *(Traci Case, 2009)*. *EPA (2006)* reported that copper corrosion could increase copper leaching in drinking water, which may cause health risks such as nausea, diarrhea, and stomach cramps, but alternative choices of corrosion inhibition can be used, like plastic pipes (e.g. CPVC, PEX) and stainless or coated steel pipes *(Juneseok Lee, 2008)*. Additionally, pipe wall materials affect chlorine decay. *Powers (2000)* and *Nguyen (2005)* determined that chlorine decay is more rapid in copper and brass pipes, but chlorine decays at a similar slow rate in PVC and lined ductile iron pipes *(Jorge Arevalo et al., 2007)*. Recent research indicated that unlined iron refers to high activity materials, while PVC, MDPE, and cement-lined ductile iron refer to low activity materials *(N.B Hallam, 2002)*.

The size of pipe systems for drinking water transmission depends on the water use conditions. Basically, the diameter of premise plumbing is relatively smaller than that of the main distribution system, resulting in a larger surface-to-volume ratio. Chlorine decay rate could vary with different sizes of pipelines *(Sharp et al., 1991)*. Furthermore, premise plumbing systems are subject to extreme temperatures *(Rushing and Edwards, 2004)*. Chemical reaction and chlorine decay rates in piped water, could increase with temperature *(Kiene et al., 1998; Wable et al., 1991)*.

The first-order equation below describes chlorine decay at various times in drinking water premise plumbing *(Wable et al., 1991; Biswas et al., 1993; Rossman et al., 1994)*:

$$dC/dt = -kc$$

where k is the chlorine decay constant (h^{-1}), and varies with different pipe properties (e.g. diameter, pipe wall material) and the conditions inside the pipe wall (e.g. deposits, water flow rates, nutrients). This equation is important for designing premise plumbing and replacing re-chlorination equipment; it can also be applied to predict the required chlorine concentration *(Andreas Richter, 2001)*.

CHAPTER 1

Effects of Service Age and Usage Pattern on Microbiological Quality of Drinking Water after Stagnation in Premise Plumbing

1.0 INTRODUCTION

The World Health Organization (WHO) defines drinking water as “suitable for human consumption and for all usual domestic purposes including personal hygiene;”, thus, making our drinking water safe to drink and not polluted has been the primary requirement for health and sustainable life. Worldwide, million of miles of drinking water distribution pipes provide treated drinking water to final customers *(USEPA)*. Ideally, treated water from treatment plants should not have changes in quality until it is consumed or used by the consumer, but in reality, substantial changes could occur with complex physical, chemical, or biological processes *(NRC, 2006).* Therefore, as the critical infrastructure that people rely on all the time, water distribution systems deserve much more attention. The U.S. Environmental Protection Agency (EPA) states that thanks to the efforts of localities and their drinking water treatment plants devoted to solving the health and safety issues from water distribution systems, potable water in the United States is treated and monitored well compared to those in third-world countries. However, what happens to the drinking water quality in the pipes at end-used sites, known as premise plumbing?

A report indicated several characteristics in premise plumbing *(NRC, 2006)*:

(1) High surface-to-volume ratio results in more leaching and permeation,

(2) Various periods of residence time,

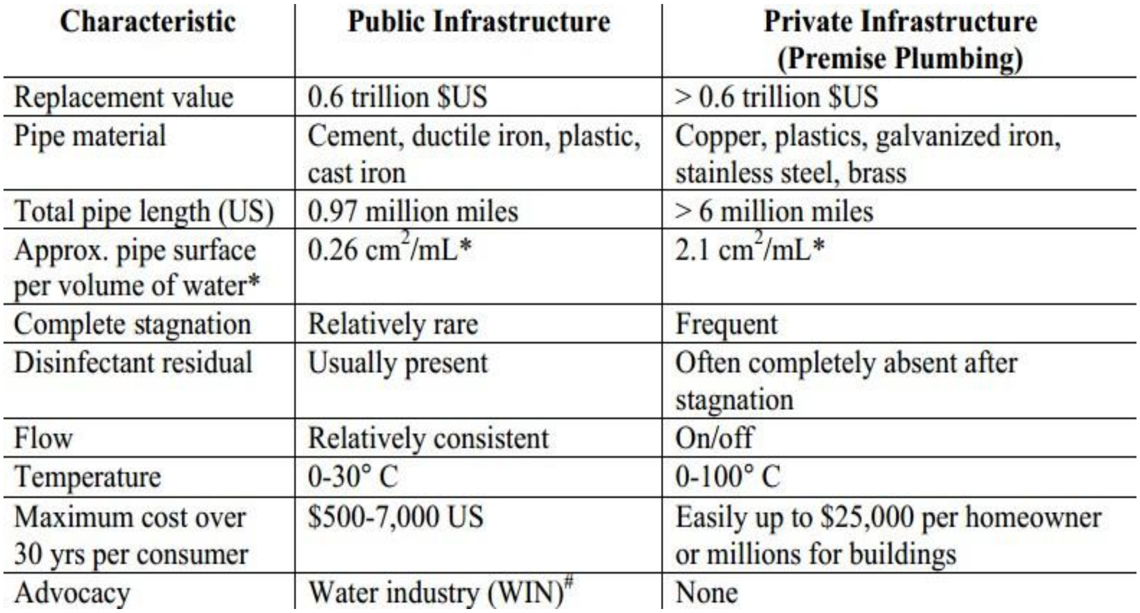
(3) Disinfection loss,

(4) Elevated temperature,

(5) Higher microbial contaminants and potential pathogen risk.

Basically, microbial re-growth could occur by excess residence time of drinking water, the time drinking water is stagnated in the premise plumbing before it flows again through the pipelines for consumer use *(Caroline Nguyen, 2008)*. Stagnant conditions of drinking water are frequent in premise plumbing (Table 1), and may represent important ecological niches for a wide variety of microbes in order to cause public health risks *(Snoeyink et al., 2006)*. Other research also indicated that bacteria can re-grow in premise plumbing to several orders of magnitude higher than that in distribution systems *(Edward et al., 2005)*. Thus, chapter 1 focuses on the microbial contamination of stagnant water in premise plumbing.

Table 1. Comparison of U.S. Public and Premise Plumbing Systems *(Nguyen, C., 2008)*



Characteristic	Public Infrastructure	Private Infrastructure (Premise Plumbing)
Replacement value	0.6 trillion $US	> 0.6 trillion $US
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*	0.26 cm^2/mL*	2.1 cm^2/mL*
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	$500-7,000 US	Easily up to $25,000 per homeowner or millions for buildings
Advocacy	Water industry (WIN)$^{\#}$	None

A large range of service ages of premise plumbing systems has been constructed in the United States, and even the pipes from the late 19th century are still used at present *(NRC 2006)*. Reports indicate that service age of premise plumbing could affect water quality *(Kelly A. Reynolds et al., 2008)*. As pipe service age increases, disinfectant loss rate increases *(Patrick Asamoah Sakyi1, 2012)* and then results in premature water stagnation, thus higher bacterial density would appear under such stationary water conditions in premise plumbing *(Maul et al. 1985; J. Wingender, 2004).* Therefore, as pipe service age increases, microbial contamination levels may increase.

In addition to service age, the service age, usage pattern of premise plumbing is another important factor for water quality. The definition of usage pattern is based on flushing frequency through the premise plumbing. Reports indicate that the change of flow rate by quickly opening or closing a faucet could wash away the tubercles and deposits on pipe inner surfaces. These contaminants may flow back and worsen water quality. However, flushing strategy, known as usage pattern, can effectively remove these undesired contaminants to keep the water clean *(Z. Michael Lahlou, 2002).* Moreover, 95% of the potential microbiological contaminations was from biofilm on the pipe surface, but higher usage patterns could flush microorganisms and reduce biofilm accumulation thereby destroying habitats for bacterial re-growth *(J. Wingender, 2004)*.

Heterotrophic plate counts (HPC) is a biological method to characterize bacteria density in order to test water quality from premise plumbing systems, particularly for tap water. Although there is no direct correlation between HPC levels and waterborne pathogens or health diseases (WTO, 2002), HPC is still a reliable method to determine the level of undesirable microbial re-growth in premise plumbing system *(Nguyen, C., 2005)*.

Overall, both pipe service age and usage pattern may greatly affect microbial contamination (HPC levels) in stagnant water. However, few researchers has investigated this linkage.

Therefore, chapter one provides an investigation on how the pipe age and usage pattern affect microbial contamination under stagnant water conditions of premise plumbing.

1.1 MATERIALS AND METHODS

1.1.1 Sampling

This research focused on the relationship between pipe service age and usage patterns on HPC levels under stagnation conditions of premise plumbing. In order to accomplish the aim of this survey, samples were collected from janitor rooms and bathrooms in various buildings of University of Tennessee, Knoxville. Table 2 below lists the stagnant water sampling sites that were listed for performing the experimental work. Every building on campus received the same treated drinking water from a traditional water treatment plant; tap water was allowed to stagnate in pipes overnight (>12 hours) in order to get rid of free chlorine residual and increase microbial counts. For each water sample, the first 30 ml of stagnant water for each site was collected by using sterile polypropylene centrifuge tubes, and then 5ml were taken out from the centrifuge tubes, Orbeco-Hellige Aqua Comparator test kits were used to quantify the chlorine residual in order to determine whether it had already been depleted (< 0.05 mg/L). All of the pipe material used in this study was copper with a nominal diameter of 0.5 inch. All samples were kept on ice in a cooler until further testing was possible, and all microbial testing was performed within 8 hours of sampling.

Table 2. Sampling Sites

Janitor Room locations	Pipe Age	Floor	Floor Capacity[a] (Person/floor)	HPC (CFU)	
				Mean	Stdev
Earth and Planetary Sciences Building	84	2	88	441.5	34.18
		3	187	273	83.79
Ferris Hall	83	3	66	437.6	36.03
		5	206	230.1	34.18
Perkins Hall	65	1	110	299.8	89.3
		2	188	134	92.69
Howard H. Baker Jr. Center	6	3	43	89.3	21.02
		2	113	62.32	26.29
James A. Haslam Business Building	4	5	160	170.2	56.41
		2	424	147.2	48.11
Min H. Kao Electrical Engineering & Computer Science	2	3	15	69.92	35.77
		1	247	50.4	30.69

Bathroom Locations	Pipe Age (yr)	Floor	Floor Capacity[b] (Person/floor)	HPC (CFU)	
				Mean	Std. Dev
Melrose Hall	113	1	6	561	46.29
		2	16	550	97.65
		3	7	568	42.52
		4	9	537	80.07
Estabrook Hall	110	1	188	384	78.94
		B	157	351	38.50
Pasqua Nuclear Engineering Building	88	2	112	272	29.82
		3	130	334	78.00
Jessie W. Harris Building	87	1	171	263	51.64
		2	220	283	36.12

Table 2. *Continued.*

		3	80	378	57.47
Earth and Planetary Sciences Building	84	1	88	324	48.86
		2	125	248	39.87
		3	187	194	36.02
		4	173	275	62.25
Ferris Hall	83	3	244	270	65.78
		4	488	190	57.97
College of Nursing	42	1	280	119	50.40
		2	297	79	22.12
		3	115	205	8.02
Student Services Building	42	1	172	151	34.86
		2	118	134	46.02
		3	60	265	61.66
		4	118	109	52.28
Stokely Management Center	40	2	63	154	50.1
		3	83	183	41.1
		4	76	157	57.8
		5	66	196	29.0
		6	85	253	65.9
		7	92	188	64.5
		M	6	361	65.93
John C Hodges Library	26	1	269	31	13.4
		2	345	28	24.3
		3	351	37	22.6
		4	198	166	53.3
		5	160	237	62.1
		6	105	284	90.2
Science and Engineering Building	16	2	202	92	28.1
		3	353	95	21.3
		4	229	60	22.7
		5	153	112	17.6
		6	90	150	42.6
		7	121	180	54.8
Howard H. Baker Jr.Center	6	1	200	97	37.7
		2	113	229	66.0
		3	43	222	24.8
		4	118	176	58.6

Table 2. *Continued.*

		5	137	154	46.5
		6	72	221	83.7
Min H. Kao Electrical Engineering & Computer Science	2	1	15	369	69.4
		2	211	170	57.0
		3	124	129	50.1
		4	237	138	52.5
		5	179	123	39.7
		6	151	165	77.6

a. Unit is person per floor which was proportional to the floor area that needed to be cleaned by janitors.
b. Unit is person per floor which was proportional to usage pattern of flooring tap in bathrooms.

1.1.2 Heterotrophic Plate Counts (HPC)

A low-nutrient media (R2A) was used since it has been determined to work well for heterotrophic bacteria in tap water systems *(Martin J. Allen etc al.,2002)*. For each water sample, 0.2mL was spread on the surface of an R2A agar plate *(Reasoner and Geldreich, 1985)*, and colony-forming units (Cfu) were counted after incubation at 28 ^{o}C for seven days.

1.1.3 Additional Water Quality Parameters

Free chlorine concentration (Orbeco-Hellige Aqua Comparator test kits with DPD method) and conductivity were measured on site. A digital thermometer was used to determine the water temperature in premise plumbing systems.

1.1.4 Statistical Analysis

JMP Pro 9.0 was primarily used for statistical analysis. Samples were separated into different categories by pipe service age and usage pattern, and then compared using an analysis of variance (ANOVA). Comparisons for all pairs using Tukey-Kramer HSD were also necessary to test the differences between groups. Significant level (p-value) was the most important indicator with $p<0.05$. Correlations (PEARSON formula from Excel) were performed to show relationships, while R^2 values and related p-values show the level of the relationships.

1.2 RESULTS AND DISCUSSIONS

1.2.1 Analysis of Water Quality In Janitor Rooms

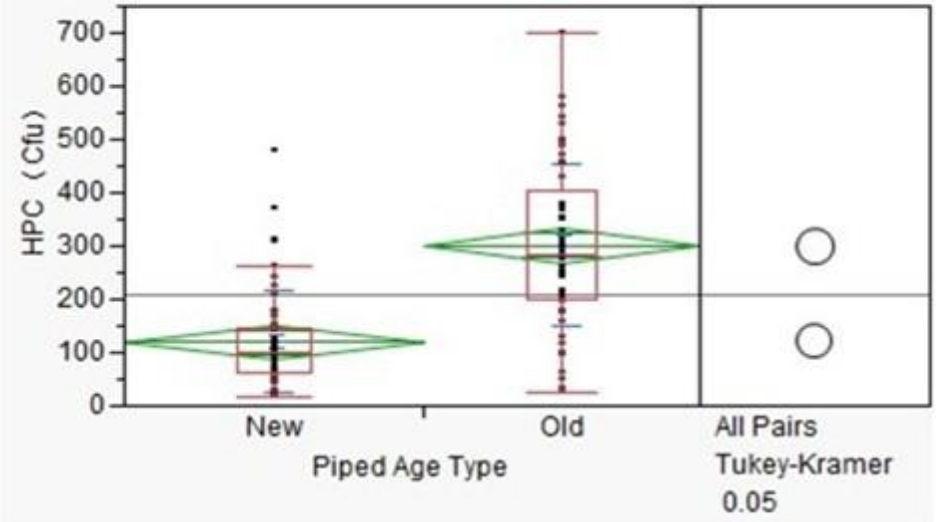


Figure 1. HPCs from Stagnant Water of Premise Plumbing in Different Aged Pipe Systems ($p<0.05$) by ANOVA

Janitor rooms were used once per day for cleaning, maintenance, and security responsibilities. It was therefore assumed that the taps in each janitor room had the same usage pattern. Stagnant water samples were consistently collected from taps of janitor rooms approximately 19hrs after their previous use. Measurements of viable heterotrophic bacteria counts (HPC) characterized general surface colonization. As pipes age, chlorine decay rate increases may result in increased microbial levels (*A.O. Al-Jasser, 2006*). In this study, the same results appeared, showing a significant difference of HPC levels between different aged pipe systems (Figure 1). Additionally, as pipe service age increased, HPC levels increased as well.

After separating the whole data by flooring samples, results showed significant differences between flooring samples in each of the older plumbing systems (Figure 2): the significant levels of HPC ranged from 0.002 to 0.019 (all $p<0.05$), but there were no significant differences between different flooring samples of HPC from newer pipes (Table 3).

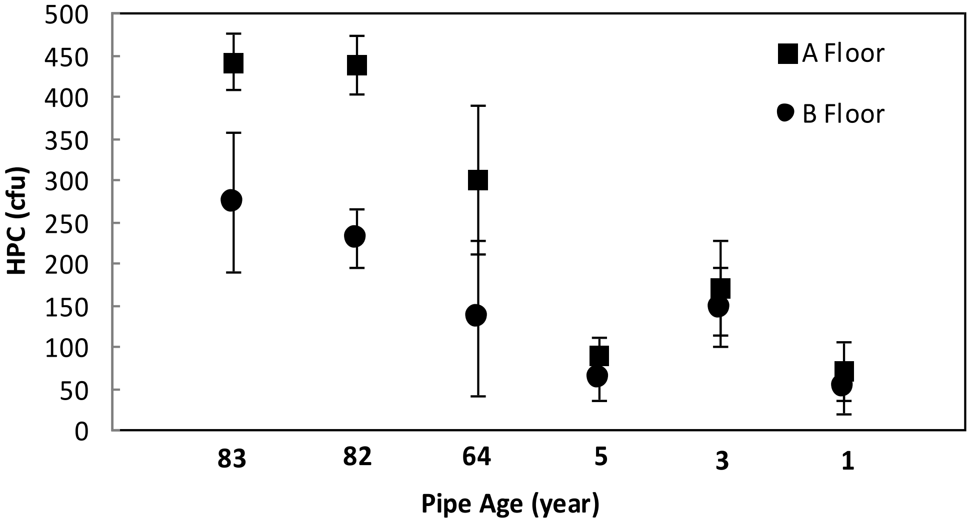


Figure 2. HPC Levels (Cfu/200ul) of Different Flooring Samples in Various Aged Janitor Rooms

Although the initial assumption was to determine that the usage pattern of janitor taps was the same (that is, different flooring samples should have similar HPC levels in each of the aged buildings), the surprising results for each of the older flooring samples may be due to the particular usage pattern in reality, like different water volumes used and flushing times kept by various behaviors of janitors. Maximum floor capacity (person/floor) was assumed to be proportional to the floor area that needed to be cleaned, known as usage pattern (Table 2). In other words, if there were more classrooms on a floor, the maximum floor capacity could increase, and then the cleaning area would therefore increase, and more water volume and longer flushing time would be required in the janitor room for that floor. Different usage patterns of pipes may affect environmental conditions of the inner plumbing, that is, greater water volume use, higher usage of taps and even longer flushing time could remove unwanted microbial contaminants or deposits inside the premise plumbing, thereby reducing the potential of bacteria re-growth and accumulation. In general, my speculation is that different and specific floor conditions may lead to different usage patterns, which affect microbial survival and plays a greater role on HPCs of older flooring samples. However, based on the comparison with older systems, specific flooring usage pattern did not affect HPCs as much in each of the newer systems (all $p>0.05$).

Furthermore, according to the correlation between usage pattern and HPCs, there was the negative value (-0.896) in older premise plumbing systems, while there were no such significant correlations (0.177) in newer systems.

Table 3. Significant Level of HPCs from Different Flooring Samples in Each Aged Premise Plumbing System

Pipe Age	Building List	Pipe Age (yr)	Floor	Floor Capacity[b] (Person/floor)	Mean	Stdev	**Sig.**[*]
Old	E & P	84	2	88	441.5	34.2	0.016
			3	187	273.0	83.8	
	F	83	3	66	437.6	36.0	0.002
			5	206	230.1	34.2	
	P	65	1	110	299.8	89.3	0.019
			2	188	134.0	92.7	
New	B	6	3	43	89.3	21.0	0.216
			2	113	62.3	26.3	
	H	4	5	160	170.2	56.4	1.000
			2	424	147.2	48.1	
	M	2	3	15	69.9	35.8	0.854
			1	247	50.4	30.7	

* Significant levels of HPC between two different flooring premise plumbing systems.

1.2.1.1 Water Quality Measurements

Conductivity describes the degree to which an aqueous solution carries an electric current. The results of this study showed that the stagnant water sampled from 83 to 84 years old aged pipes had relative high conductivity (Figure 3). Some researches have indicated that the high conductivity levels in older pipes may be due to high levels of metal leached into the water *(Brian Oram ect.al, 2002)*

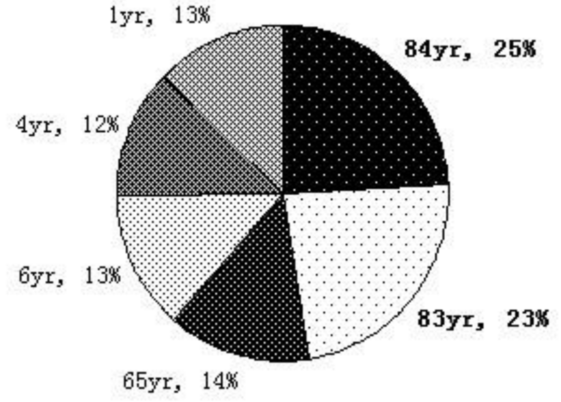


Figure 3. Conductivity of Each Aged Building.
"%" was the comparison of conductivity between different aged buildings, while "yr" means how old was the pipe system

1.2.2 Analysis of Water Quality in Bathrooms

Besides the effect of service age on stagnant water quality, flushing is another factor that affects the potential risk of microbial contamination and re-growth, especially for biofilm accumulation in premise plumbing systems *(Benoit Barbeau, 2005).* Bathrooms were a better choice to collect water samples because the bathroom taps in each building are much more frequently used than the faucets for custodial use; thus, the usage patterns are highly variable for each floor of each building. From these experimental sites, the study focused on how usage pattern and service age affect the HPCs of stagnant water in premise plumbing of bathrooms. Bathroom flooring samples (Table 1-Bathroom sites) with their own usage patterns were collected, and the maximum floor capacity (person/floor) was assumed to be proportional to the usage pattern for each flooring bathroom.

In order to specifically identify the impact of usage pattern on water quality, the premise plumbing system samples in this study were separated into three age categories: Old (range from 113 years to 83 years); intermediate (range from 42 years to 26 years) and new (range from 16 years to 2 years). By applying statistical analysis for correlation, results indicated that there was a significant negative correlation between usage pattern and HPCs as shown in Table 4, which shows that as usage pattern increased, levels of HPC decreased, which indicated a better water quality.

Table 4. Correlation between Usage Pattern and HPCs under Different Aged Pipes.

Year Category	**Usage Pattern vs. HPC**
New	-0.67
Intermediate	-0.70
Old	-0.64

Moreover, results of the ANOVA test and post-hoc analysis showed that there was no correlation between the age of the pipe systems and the level of HPC, but there was a significant difference in the levels of HPC between the low and high usage category (all $p<0.05$); Additionally, in newer pipe systems, usage pattern has a more significant impact on HPCs (all $p< 0.05$) (Figure 4).

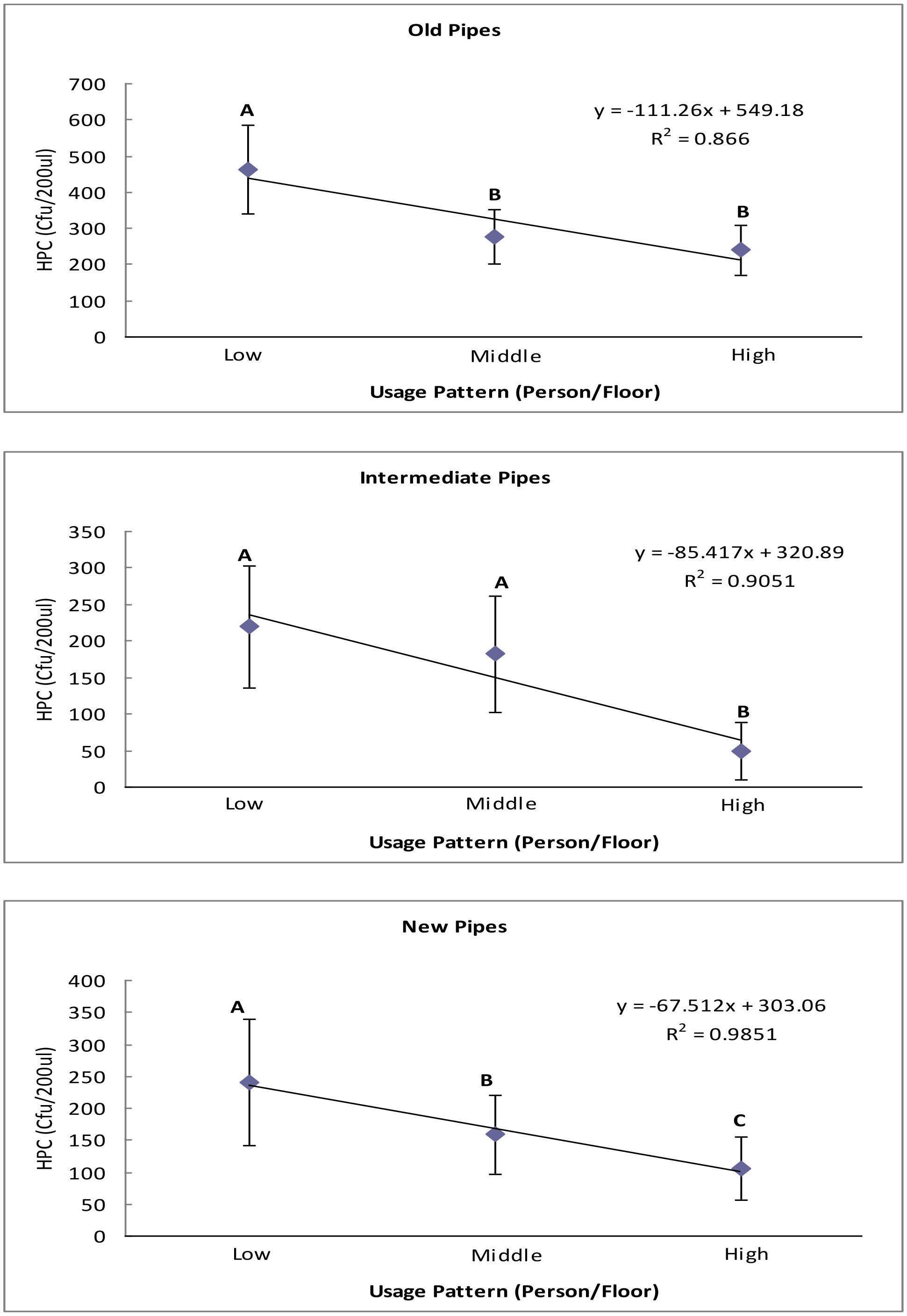


Figure 4. Effect of Usage Pattern on HPCs under Different Aged Premise Plumbing.

A,B,C Symbol: Comparison of significant level between two groups at $p<0.05$. There was a significant difference between different symbols ($P<0.05$), while no significant difference between same symbols.

In order to investigate the effect of pipe service age on levels of HPCs, bathroom samples were separated into another three categories: low-usage pattern pipes, middle-usage pattern pipes and high-usage pattern pipes. Effect of pipe service age on HPCs could be analyzed separately for each category. Figure 5 shows the significance difference of HPCs under different usage pattern categories. No matter what the usage pattern was, old aged pipes had the most viable microbes inside, thus having the greatest impact on HPC than other aged pipes, which was indicated by $p<0.05$.

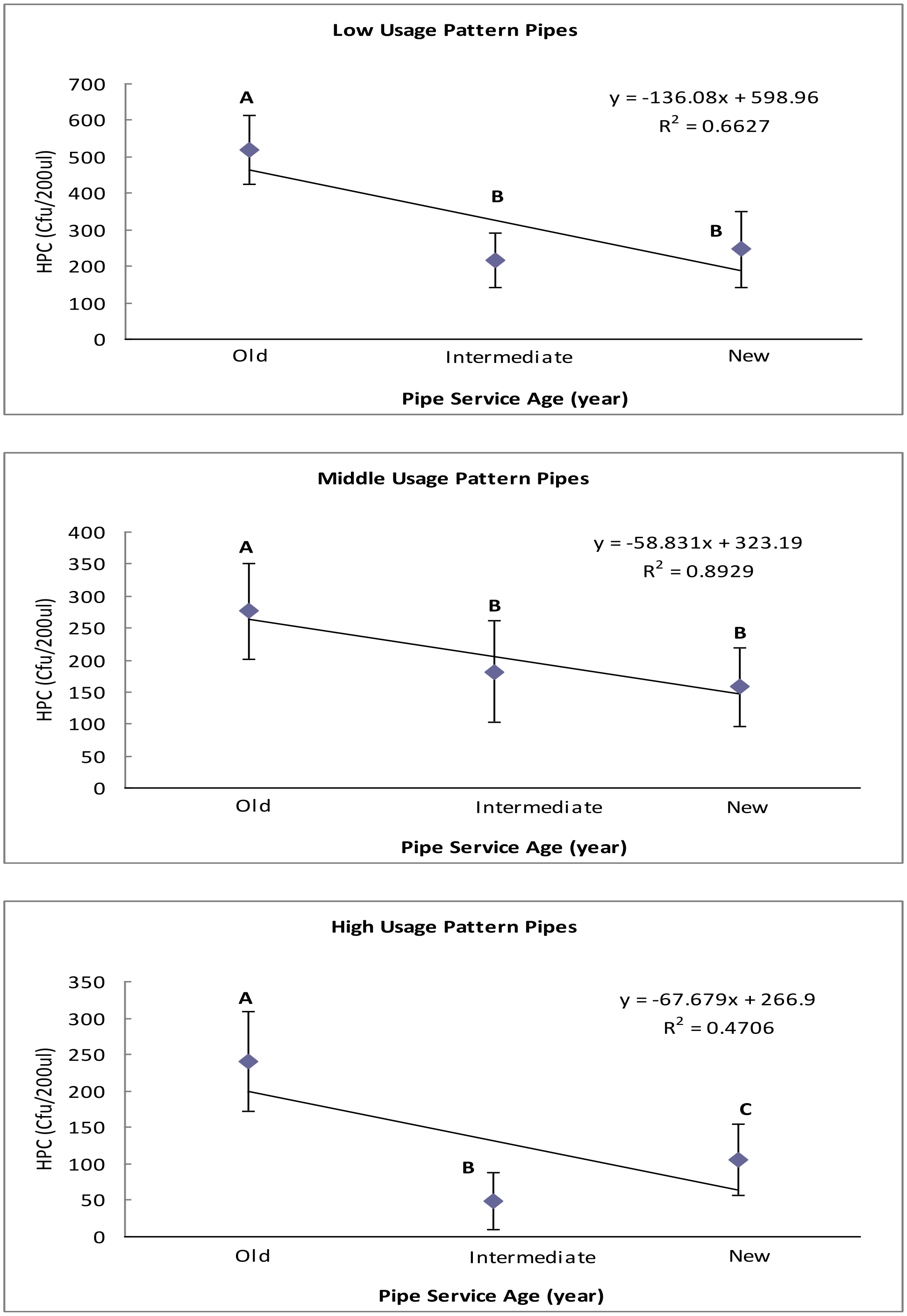


Figure 5. Effect of Pipe Service Age on HPCs under Different Pipe Usage Patterns A,B,C Symbol: Comparison of significant level between two groups at $p<0.05$. There was a significant difference between different symbols ($p<0.05$), but no significant difference between same symbols ($p>0.05$).

Furthermore, based on the correlation analysis (Table 5), there was a positive correlation between service age and HPCs. As pipe service age increased, the HPCs increased; especially in the low usage category, the correlation was more significant (0.72).

Table 5. Correlation between Pipe Service Age and HPC Level under Different Usage Patterns.

Usage Pattern Category	**Pipe Service Age (Year) vs. HPC**
Low Usage pattern System	0.72
Middle Usage pattern System	0.55
High Usage pattern System	0.58

1.3 CONCLUSIONS

(1) The service age of premise plumbing systems is positively and significantly correlated to the concentration of microorganisms in stagnant drinking water.

(2) Usage pattern is another factor contributing to microbial contamination, with systems experiencing lower levels of water usage pattern exhibiting greater microbial contamination than those having greater water usage pattern, potentially due to the prolonged periods of stagnation and subsequent greater disinfectant decay rate.

1.4. SUGGESTIONS

(1) New pipes are better constructed to loop end points in order to prevent stagnant water from occurring *(EPA, 2007)*.

(2) Regular flush plumbing with fresh water can keep disinfectant levels constant and reduce the potential of microorganism re-growth and accumulation, and even affect the odor and taste issues *(Caroline Nguyen, 2008)*.

CHAPTER 2

Impact of Pipe Material and Deposits on Chlorine Decay Rate

2.1 INTRODUCTION

Water is a necessary and essential source for human, animals, and even vegetables, therefore its microbial and pathogenic contaminations should be eliminated to avoid conditions that lead to health risks. In order to maintain potable water quality, disinfectant is added to water, which then enters the drinking water transmission and distribution system*s (Benoit Barbeau, 2005). M*ost waterborne pathogens and microorganisms are inactivated as a result of the disinfectant. Disinfection can also improve drinking water quality by reducing taste, odor, and color, and leading to the oxidation of ferrous iron, manganese, hydrogen sulphide and cyanides *(Pierce, R.C. 1978).*

Historically, water-soluble chlorine was discovered in reactive and corrosive chlorine gas (*Carl Wihelm Scheele, 1774)*, which was applied to bleach for paper, vegetables, and flowers. Today, based on the stabilization, effectiveness, and low costs, chlorine is broadly and commonly applied to disinfect drinking water with an excess of 0.2mg/L concentration (*Clark, R.M., Coyle, J.A., 1990*). However, excess chlorination is not appropriate due to by-products such as trihalomethanes, which are considered carcinogenic. The formation mechanism of water-soluble chlorine is shown below *(website-disinfection)*:

$$Cl_2 + H_2O = HOCl + H^+ + Cl^-$$

$$HOCl + H_2O = H_3O^+ + OCl^-$$

$$OCl^- = Cl^- + O$$

Higher reactive hypochlorous acid (HOCl) and hypochlorite ions (OCl^-) are considered as free chlorine; strong oxidizing oxygen atoms replace their atoms to other compounds like enzymes of bacteria, which results in bacterial break down. Thus free chlorine becomes a good measureable and effective method to estimate the potability of drinking water and waterborne microorganisms level *(website-disinfection)*.

Premise plumbing systems have prolonged residence times, more stagnant time and elevated temperatures resulting in worse adverse effects on drinking water quality reaching consumers in comparison with the main distribution system. Additionally, chlorine decay more rapidly in premise plumbing, influenced by a range of physicochemical and biological reasons, such as reactions with pipe materials and organic/inorganic nutrients of drinking water, biofilm, tubercles and corrosion by-products produced on the pipe wall surface *(Wabl,O, 1991;* DiGiano and Zhang, 2005; *N.B. Hallasm et al., 2002)*.

It would be a useful tool for designing and operating premise plumbing to model the dynamic fate of the free chlorine of drinking water. The decay has been expressed by a first order reaction for both the wall and bulk reactions *(Chambers et al., 1995)*, which shows that free chlorine concentration changes with resident times in drinking water premise plumbing systems for drinking water.

$$dC/dt = -kC$$

$$\text{or } C_t = C_0 \exp(-kt)$$

k is the first-order decay constant (h^{-1}) and $k = k_b + k_w$, where k_b represents the bulk first-order chlorine constant (h^{-1}) and k_w represents the wall first-order chlorine decay constant (h^{-1}). In other words, the total chlorine decay rate involves either reaction with water compounds (bulk decay) or pipe walls (wall decay). However, wall decay is considered as a dominant mechanism compared with bulk decay within drinking water *(Huang J, 2008).*

Drinking water premise plumbing systems are constructed with various types and ages of pipelines, ranging from cast iron pipes during the 19th century to ductile iron pipe and finally to plastic pipes installed in the 1970s and still in use *(NRC, 2006).* Today, premise plumbing systems involve a variety of materials including copper, galvanized iron, plastics, brass, lead and stainless steel. In order to maintain potable water quality, disinfectant is added to the treated water. Once disinfectants decay occurs, it can cause a series of water quality degradation. Researchers have investigated the reaction between free chlorine and various pipe materials of premise plumbing (Clark et al., 1994); Powers (2000) and Nguyen (2005) also determined that chlorine decays more rapidly in copper and brass pipelines. Moreover, recent research indicates that unlined iron are considered high activity materials, while PVC, MDPE and cement-lined ductile iron are considered low activity materials *(N.B Hallam, 2002)*. However, the effects of pipe materials on disinfectant decay have often been underestimated by drinking water treatment plants *(H. J. Singleton, 1989)*.

Premise plumbing itself is not an independent system that could interact with the pipes' inner aqueous environment or pipe wall. During the interaction process, undesired deposits could be produced *(Heryong Jung, 2009)* by the physicochemical and biological reactions with particulate matter, dissolved oxygen, chlorine, sulphates from bulk water body and pipe inner wall surfaces. Recent research has determined the primary deposits in drinking water premise plumbing include minerals, organic matters, corrosion products, and biofilm *(Vikas Chawla, 2012)*. Deposits have adverse effects not only for flow of water transmission *(S.A. Imran, 2005)* but also on water quality. Many researchers have investigated the effect of deposits on drinking water quality, for instance, odor, taste, and color issues could be produced due to the release of corrosion products *(Sarin et al., 2004; Imran et al., 2005)*. Microbial contamination could result from reactions between biofilm, humid substances, and iron oxide *(Lechevallier et al., 1987; Zacheus et al., 2001)*. Furthermore, *Gauthier et al. (2001)* indicated that deposits can result in severe reductions in disinfectants of drinking water and even reduce free chlorine residual *(Lehtola et al., 2004)*; deposits have also been found to reduce chlorine decay and provide nutrients for bacteria growth

(Gauthier et al. 1999). However, few studies have investigated the effects of deposits on water quality in pipelines of different materials, especially for disinfectant (free chlorine) decay problems.

2.2 OBJECTIVES OF RESEARCH:

(1) Characterize the effects of pipe materials on free chlorine decay,

(2) Characterize the influence of deposits in the pipe on chlorine decay rate.

2.3 MATERIALS AND METHODS

The experimental system includes twelve U-shaped plumbing pipes with a nominal diameter of 0.5 inch and length of 60 inches. Different pipe materials contain galvanized iron, copper, and PVC (four times repetition). Each pipe system was equipped with a submersible pump in a closed reservoir of 15-L drinking water. These pipelines had already been in operation (for prior biofilm experiments) for around three years using tap water before this study and thus there was deposits accumulation on the inside surface of the pipes (*Yan Zhang, 2012*).

After all of the pipes were filled with tap water and equilibrated around 24 h at room temperature (22℃), the pipes were rinsed with DI water before being filled with tap water again for measuring the chlorine decay rate. Tap water was filled in pipes from the same faucet with the same flow rate (adjusted by ball valve), and then the pipelines were placed at room temperature to ensure chlorine consumption. For each sample, 5ml of stagnant water were collected at certain time intervals for testing free chlorine until chlorine concentration dropped below 0.05mg/L using the Orbeco-Hellige Aqua Comparator test kits with DPD colorimetric method. A digital thermometer was used to measure the initial water temperature, which was 16℃. By applying the first-order decay equation, the natural log of chlorine concentration was plotted against water stagnation time. The chlorine decay constant (k) was determined by the slope of linear regression line (Figure 6 as an example)

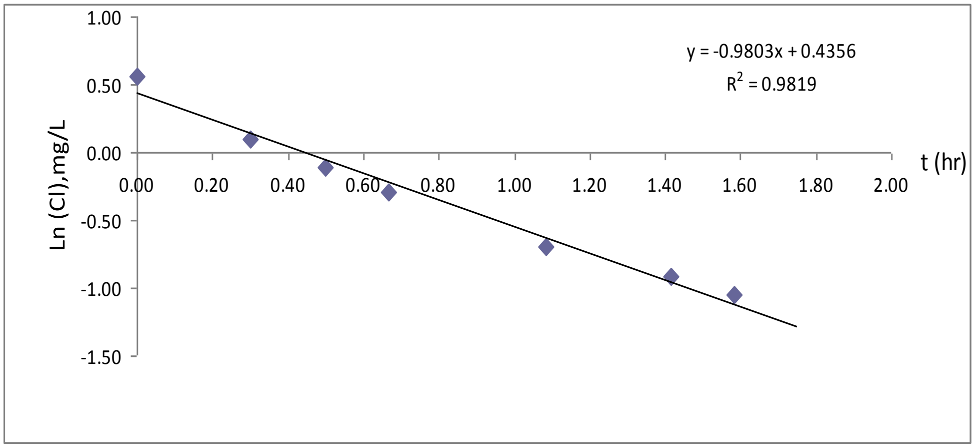


Figure 6. Chlorine Decay Over Time in Copper Pipe (As an example)
The k-value was 0.9803 hr^{-1} with the R^2 of 0.9819.

Water-soluble free chlorine solution (10% domestic bleach) was filled into all twelve pipelines, and the pipes were hand-shaken in the same direction 5 min and then the diluted free chlorine fluid inside the pipe systems were placed at room temperature overnight (>8h). Then the pipes were flushed with tap water for 30 min to clean the inside of the pipes, which was followed by DI water rinsing. The DI water rinsing consists of filling the pipe with tap water again and measuring chlorine decay in the series of pipelines based on DPD method by Orbeco-Hellige Aqua Comparator test kits. Triplicate measurements were needed for each sample at each time to reduce k-value bias. In the end, pipes were rinsed with DI water and stored inside tap water to protect the inner pipe environment. Chlorine decay rate was measured again after half a year.

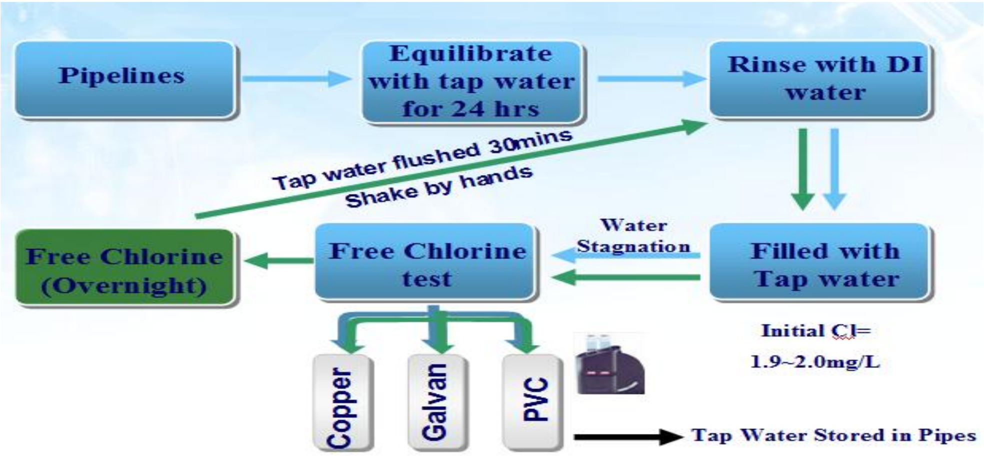


Figure 7. Flow Chart of The Methods for the Effects of Pipe Materials and Deposits on Chlorine Decay.

2.4 RESULTS AND DISCUSSION

2.4.1Effect of Pipe Materials on Chlorine Decay Rate

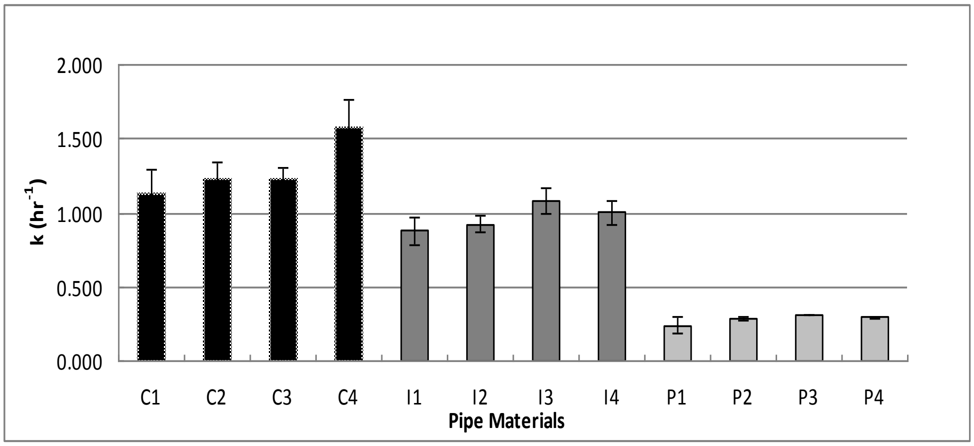


Figure 8. Effects of Pipe Materials on Chlorine Decay Rate.

*C, I, and P represent copper, galvanized iron and PVC pipelines, respectively.

Chlorine decay rates varied from 0.24 hr^{-1} to 1.57 hr^{-1} for different pipe materials. Average chlorine decay rate was greatest in copper pipes as shown in Figure 7. Copper and galvanized pipes exhibited relatively high reactivity, while PVC was a relatively non-reactive pipe material. Compared with other work, k-value might vary with the conditions and characteristics of the pipe itself *(Caroline K. Nguyen, 2005)*.

2.4.2 Effect of Free Chlorine on Chlorine Decay Rate

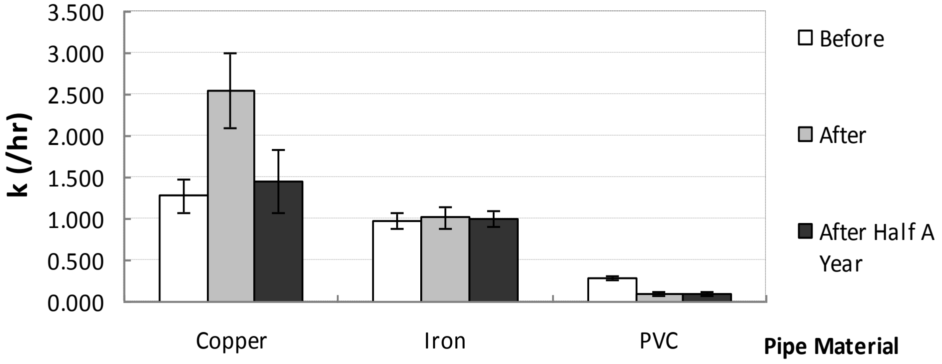


Figure 9. Effects of Free Chlorine on Chlorine Decay Rate in Different Pipe Materials.

After using free chlorine to remove deposits in the pipeline systems, chlorine decay rate k increased for copper pipes from 1.274/h to 2.557/h, and decreased for PVC pipes, but there was no significant difference for galvanized pipelines (Figure 8). Other researchers have indicated that deposits surely have effects on chlorine decay rate. When deposits were present in the pipe, chlorine decay was slower than when deposits were not present *(Domanska, 2011)*. Based on my study, this phenomenon occurred in copper pipes; that is, after cleaning away the water-pipe deposits, chlorine decay rate increased significantly. Furthermore, after stagnant water was stored in pipes for half a year, the chlorine decay rate of copper pipe dropped to 1.455 hr^{-1}, while other materials did not show significant changes. Formation of deposits is speculated but more future research is needed to clarify this speculation.

2.4.3 Effect of Pipe Materials on Water pH

Measurement of the pH of the stagnant water over time following water stagnation in galvanized iron pipes determined that PH increased more rapidly than in other material pipes. Some work states that iron releasing is a large factor in rapidly increasing pH *(Robert M, 1999)*.

2.5 CONCLUSIONS

(1) According to pipe material characteristics, the order from fastest chlorine decay rate to lowest was as follows: Copper pipes> Galvanized iron pipes> PVC pipes;

(2) Deposits slow down chlorine decay, while chlorine decay is increased without deposits. This effect occurred mainly in copper pipes;

(3) pH increased rapidly following water stagnation in galvanized iron pipes.

2.6 SUGGESTIONS:

Appropriate pipe materials of premise plumbing need to be designed and installed to reduce adverse water quality impacts including releasing unwanted metals and producing deposits.

CHAPTER 3

Models of Chlorine Decay Rates of Stagnant Water in Premise Plumbing of Drinking Water Distribution System

3.1 INTRODUCTION

Disinfectant is added into treated water at the drinking water treatment plant and transferred to distribution systems, but the concentration of disinfectants does not remain constant especially during the transmission to the consumer tap—which engineers refer to as premise plumbing systems. As a widely used disinfectant, free chlorine disappears gradually due to a variety of reasons and thus increases the possibility of microbial contamination. This phenomenon usually occurs in premise plumbing systems with longer water residence time *(Vieira P, 2004).* Chlorine wall decay constant (k) is harder to determine as a function of a variety of pipe properties: pipe service age, deposits, usage pattern, pipe material, diameter, and ambient elevated temperature. Prior studies have already stated the effects of service age, deposits, and usage pattern, thus chapter three will focus on the effect of pipe material, diameter, and ambient elevated temperature. For modeling aims, these factors should be studied separately in order to quantify the effect of each decay mechanisms correctly. The main objective was to quantitatively investigate the relationship between the chlorine consumption rate mechanism and the effects of pipe materials, diameter, and temperature.

3.2 MATERIALS AND METHODS

The pilot pipelines were bought from a *Home Depot* store in Knoxville, TN and were established in the laboratory condition. The performance of different pipe materials was tested with new galvanized steel, copper, and PVC. For each material pipe, different pipe diameters (0.5 inch & 0.75 inch) were set up with total length of 24 inches. For effects of temperature, incubation places under different temperature condition were applied (21℃, 35℃, 5℃). The stoppers were equipped with a manual control to open and close in order to collect water samples for measuring free chlorine concentration at different time intervals. A color-wheel test kit was used to measure chlorine concentration at the inlet of pipes based on color changes. According to the reaction with DPD (N,N diethyl-p-phenylene diamine), free chlorine could cause the color to change from clear to pink. Free chlorine reading ranges from 0-3.5mg/L (0-3.5ppm). Duplicated water samples were collected at each time interval to reduce the bias of chlorine measurement.

3.3 RESULTS AND DISCUSSIONS

3.3.1 Wall Chlorine Decay Mechanism

Wall chlorine decay is the reaction between chlorine and the pipe wall material. When water flows through pipes in water distribution systems, dissolved compounds can be transported to the pipe wall and react with the products on the wall (corrosion by-products or biofilm) or just react with the pipe wall itself. Previous work determined that chlorine decay in water through pipe systems is driven by first order kinetic ($dC/dt = -kC$). Wall decay constant k is a function of pipe features, like pipe materials, pipe service age, and diameter.

3.3.2 Effect of Pipe Diameter

A Bulk decay model with boundary conditions, which expresses the chlorine decay mechanism by the reaction with pipe wall materials, was used and is shown below (*Wable et al., 1991*). Chlorine decay rate could be influenced by the whole reactive area of the wall and the rate of mass transfer between the drinking water and the pipe wall.

$$\frac{\partial C}{\partial r} = -A_2 C$$

This study was performed on two different pipe diameters (0.5 inch and 0.75 inch), using three different pipe materials (copper, galvanized steel and PVC) at room temperature. The results showed that as pipe diameter increased, the decay rate constant k decreased, because smaller pipes have larger surface to volume ratios than those of larger diameter pipes (*Risala A. Mohammed, 2003*). From this bulk decay model, we can also see that the size of the wall surface does impact the chlorine decay rate.

$$R = \frac{2\pi r l}{\pi r^2 l} = \frac{4}{d}$$

.

where R is the specific area contact rate which is equal to the surface area/volume, and d is the inner diameter of pipes.

Table 6. Wall Chlorine Decay Rate With Different Pipe Diameters for Galvanized Steel, Copper and PVC Pipeline Systems (h^{-1}). ($R^2 > 0.98$)

Pipe Materials	**Diameter = 0.5 in**	**Diameter = 0.75 in**
Galvanized Steel	13.881	1.53
Copper	0.705	0.69
PVC	0.177	0.0133

3.3.3 Effects of Pipe Materials

Chlorine decay can be affected by various factors, like biofilm, deposits, and pipe materials. Premise plumbing consists of two categories: synthetic and metallic pipes. Biofilm and deposit growth is common for both categories, but chemical reactions with pipe wall materials are not *(Jorge M. Arevalo, 2007)*. Previous work has stated that synthetic pipes include PVC, polyethylene, polypropylene, and cement-lined iron *(Kiene et al., 1998)*, while metallic pipes include copper and unlined cast iron pipes. Chlorine decay rate is highly dependent on the characteristics of different groups *(Jorge M. Arevalo, 2007).*

New copper, galvanized steel, and PVC pipelines with 0.5 inch of diameter were used for this study. As shown in Table 7, the results determined that the rate of chlorine decay rate was lowest in PVC pipes in which the material has low chlorine demand. However, chlorine consumption decreased most rapidly in the galvanized steel pipelines. The results indicated that the chlorine decay of drinking water in copper pipe was almost completed in 1.8 hrs, 0.25 hr in galvanized steel pipes and 21 hrs in PVC pipes. The k-value for galvanized steel pipe was 18 times higher than copper, and k-value of copper was 44 times higher than PVC. Thus, chlorine consumption rate was significantly influenced by pipe wall materials. Previous work indicated that the rapid chlorine decay rate of galvanized steel was due to the reaction between chlorine and metallic ions which are released from galvanized steel pipe wall.

Table 7. Effect of Pipe Materials on Free Chlorine Consumption Rate

	Galvanized Steel	**Copper**	**PVC**
K-value (h^{-1})	12.813	0.696	0.01585
Standard Deviation	0.43	0.11	0.002

3.3.4 Temperature Accelerated Chlorine Decay Rate

Chlorine decay rate increased as temperature increased *(Powell et al., 2000; Jadas-Hécart et al, 1992)*. Based on the Van't Hoff-Arrhenius equation shown below, the chlorine decay constant could be determined at operation temperature, and K_T could also be calculated from a base rate decay constant at a base temperature (20℃).

$K_T = A \times \exp^{(-E/RT)}$

$K_T = K_{20} \times \alpha^{(T-20)}$

where K_T = dissipation constant at T(℃)

A= Constant

E= Activation energy

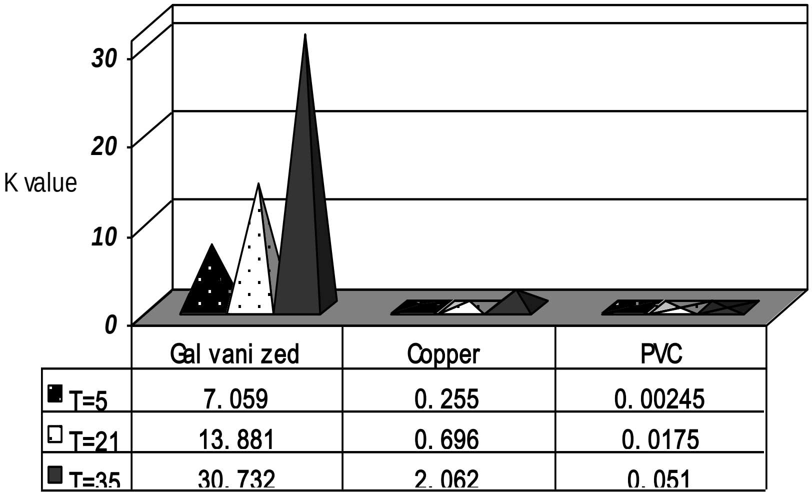
R= Ideal gas law constant

T= Temperature

K_{20}=dissipation constant at 20 ℃

α= temperature correction factor

Under different temperature treatments, chlorine decay rate changed significantly. As the temperature increased, chlorine decay rate increased (*Powell J.C, 2000*) (Figure 10). The effect was more noticeable in galvanized steel pipes.



	Galvanized	Copper	PVC
■ T=5	7.059	0.255	0.00245
□ T=21	13.881	0.696	0.0175
■ T=35	30.732	2.062	0.051

Figure 10. Effects of Temperature on Chlorine Decay Rate for Different Pipe System Materials. (Temperature was applied under 5℃, 21℃ and 35℃).

3.4 CONCLUSIONS:

(1) Free chlorine consumption in water distribution systems followed the first-order model with respect to initial chlorine concentration;

(2) When new copper, galvanized steel, and PVC pipelines were applied, free chlorine decay rate was slowest in PVC pipe systems, and was significantly faster in metallic pipe, especially in iron-based pipes;

(3) Higher temperature accelerates chlorine decay rate, while lower temperatures decrease free chlorine decay rate;

(4) Pipe diameter affects the chlorine decay rate, such that as the diameter decreased, decay rate increased.

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