

Does the onset of nitrification equally impact in decaying chloramine?

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ABSTRACT

Accelerated chloramine decay is normally observed after the onset of nitrification in the chloraminated water distribution systems. However, it is unknown whether the onset of nitrification equally impacts in decaying chloramine in different water distribution systems. To compare the impact of nitrification on chloramine decay, bulk water samples collected from the three distribution systems were tested. After the onset of nitrification, different chloramine decay rates were observed. Total decay coefficients of chloramine increased by 4–10 times in the samples obtained from Sydney Water Distribution System (SWDS) and lab-scale system whereas the decay rates increased by only 3–3.5 times in the samples obtained from Goldfields and Agricultural Water Supply System (GAWSS) after the onset of nitrification. The chloramine decay rate increased with ammonia drop rate, but the other mechanisms could not be ruled out. If chloramine residuals have to be controlled not only nitrification but also other mechanisms should be understood and controlled.

Keywords: Chloramine; Chemical decay; Microbial decay; Mild nitrification; Severe nitrification

1. Introduction

Chloramine is often used as a secondary disinfectant in water utilities to maintain a longer lasting residual and/or a reduction in the formation of regulated chlorinated disinfection by-products. The dose of chloramine for a water distribution system is decided based on water retention time, temperature and treated water quality. For example, about 1.7 mg/L of chloramine is dosed in Sydney Water distribution system (SWDS), Australia [1,2] whereas about 3.7 mg/L of chloramine is dosed in the Goldfields and Agricultural Water Supply System (GAWSS), Western Australia [3].

Stability of chloramine presents some challenges for water utilities, especially in warmer climates. Besides auto-decomposition (self-decay) and chemical decay (due to reactions with chloramine demanding compounds such as nitrite), the microbial decay of chloramine has been noted [1,4]. Microbial activities especially nitrification accelerates

chloramine decay chemically and microbiologically [1,5]. Nitrification is a two-step microbial process. Ammonia is initially converted to nitrite by ammonia-oxidizing bacteria (AOB) and then nitrite is converted to nitrate by nitrite-oxidizing bacteria (NOB). Nitrification occurs over a wide range of pH (6.5–10.0) and at temperatures above 15°C, but it can also occur at low temperatures [6]. Growth rates of nitrifiers are normally controlled by free ammonia concentration, temperature, pH, light, dissolved oxygen and chloramine residuals [7].

Nitrification has been observed more frequently at low chloramine residuals (<1.0 mg/L) [8–11]. Bal Krishna et al. [3] observed the onset of nitrification in the lab-scale system when chloramine residual dropped below 0.65 mg/L. Chloramine residuals of 1.0–2.0 mg/L should be sufficient to control nitrifiers [10], but once nitrification set in, it is very difficult to control even by increasing chloramine residual up to 8.0 mg/L [11]. Moreover, to stop nitrification through breakpoint chlorination requires a significant amount of total chlorine [12].

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In a previous study, Sathasivan et al [2] analyzed the behavior of 55 bulk water samples containing nitrite concentration < 0.02 mg-N/L, obtained from the SWDS, Australia. At the initial stage, chloramine was reasonably stable (low chloramine decay rate) and nitrite level was < 0.01 mg-N/L. This phase was termed as mild nitrification. When chloramine residuals dropped below about 0.5 mg/L, a sharp drop in chloramine and ammonia residuals were noted. In this phase, nitrite residual was > 0.1 mg-N/L and total chlorine decay was about an order higher than that noted in a mildly nitrifying phase and this phase was termed as a severely nitrifying phase. However, it is yet known whether similar behaviour after the onset of nitrification is common in other chloraminated systems and what controls such decay across the distribution system. Thus, the goal of this work was to investigate chloramine decay characteristics in the bulk water samples based on the nitrification conditions (mild and severe). Samples obtained from the two full- and one lab-scale chloraminated systems were investigated.

2. Materials and methods

2.1. Bulk water sampling details

Samples were collected from the two full- and one lab-scale distribution systems having different chloramination practice in terms of initially dosed chloramine residuals.

2.1.1. Lab-scale system

The lab-scale system set up at Civil Engineering Laboratory in Curtin University, Western Australia was employed to collect the bulk water samples. The lab-scale system consists of five reactors made of high-density polyethylene (HDPE). All the reactors were connected in series by HDPE pipe. The lab-scale system was operated by feeding surface water after maintaining chloramine residuals of 2.5 mg/L (chlorine to ammonia ratio 4.5:1) and pH 8.0. Feed water chloramine residual 2.5 mg/L was maintained with an aim of achieving various nitrification conditions (no, mild and severe) along the reactor system which normally occur in the full-scale water distribution systems. Moreover, the residual maintained in the feed water remains within the range of 1.7–3.7 mg/L chloramine which is maintained at the initial chloramination points of the Australian water distribution systems [2,3]. Dissolved organic carbon (DOC) level of feed water was 2.5–3.3 mg/L. Everyday 20 L water was continuously fed and water volume was maintained constant to gain retention time 20 ± 2 h in each reactor. Water temperature was maintained at 20.0–23.0°C along the reactors. Details of the lab-scale system set up and operation is given in Bal Krishna and Sathasivan [13]. After nitrifying conditions (no, mild and severe) have been confirmed based on nitrite and chlorine decay coefficients, bulk water samples were collected from the reactors and experiments were conducted.

2.1.2. Sydney water distribution system, SDWS

Raw water was obtained from large surface water storages and was treated by coagulation, flocculation, filtration,

followed by chlorination and ammoniation before supplied into the system. The treated water quality meets the following criteria: turbidity < 0.1 NTU, chloramine 1.5–1.7 mg/L (as total chlorine), DOC 3.3 ± 0.4 mg/L, pH 8.0 ± 0.2 and chlorine to ammonia mass ratio about 4. Water temperature in the system varies between 12 and 25°C, seasonally. Nitrification is usually noticed during December to March (summer) and later in April and May (autumn). Details of SWDS, Australia, and sampling method are provided in Sathasivan et al. [2].

2.1.3. Goldfields and agricultural water supply system, GAWSS

The GAWSS is located east of Perth, Western Australia. The main conduit transfers water from Mundaring Weir, Perth to Goldfields of Western Australia over a distance of approximately 560 km. The system is split into several extension areas, which branches from the main pipe supply water to farmlands and smaller towns. Primary disinfection takes place at Mundaring pump station. Chlorine gas is fed straight into the main pipe via a diffuser to achieve a total chlorine residual of 3.7 mg/L (chlorine to ammonia ratio ~4.5:1). Then within a few seconds, ammonia solution (20% w/v) is dosed downstream, directly into the pipe to form chloramine. The targeted free-ammonia residual is < 0.2 mg-N/L [3]. Water samples were collected from one of the branches of the GAWSS in summer (February and March) and in winter (August) and sampling method are detailed in Bal Krishna et al. [3].

2.2. Determination of the onset of nitrification

Based on the changes in total chlorine, nitrite and total ammoniacal nitrogen (TAN; summation of ammonia-nitrogen, ammonium-nitrogen, and nitrogen associated in chloramine) concentrations in the bulk water samples, nitrification stages could be divided into two phases; before and after the onset of nitrification. In the previous study [2], after the onset of nitrification sudden increase in nitrite and decrease in chloramine and ammonia concentrations were noticed. However, most samples from other chloraminated systems such as full-scale system [3] did not show increased chloramine decay rates, a specific requirement for a sign of severe nitrification along with high nitrite levels. To account these cases, the onset of severe nitrification was defined as a drop in TAN level and an increase of nitrite levels more than the experimental error from an initially measured value. Moreover, the first order kinetics was employed to determine the TAN drop rate coefficient after the onset of nitrification.

2.3. Microbial decay factor (F_m)

To quantify the role of microbes and chemicals in decaying chloramine F_m related parameters were determined before and after the onset of nitrification. Determining F_m involved four steps: sample preparation, incubation, monitoring of chloramine residuals and estimating the decay coefficient from the resulting data. Sample preparation involved splitting the sample into two sub-samples. The first sub-sample was not processed whereas the sec-

ond sub-sample was processed (filtered through a 0.2 μm polycarbonate membrane filter) to remove microbes. Both sub-samples were then incubated at a constant temperature (20°C) in a water bath covered by a lid. For each sub-sample, total chlorine was monitored over time and the decay coefficients were estimated by exponential regression. The processed sample provides the chemical decay coefficient (k_c) whereas unprocessed sample provides the total decay coefficient (k_t). The difference between k_t and k_c is the microbial decay coefficient (k_m). The F_m is the ratio between k_m and k_c . A detailed method is given elsewhere [1].

2.4. Analytical methods

The Aquakem 200, a high precision wet chemistry automated analyzer, was employed to measure TAN and nitrite concentrations for the samples collected from the lab-scale system and GAWSS. TAN was measured spectrophotometrically according to the methods described in EPA [14]. Nitrite was measured using the sulphanilamide method (4500-NO₂-B) [15]. Details of measurement methods for TAN and nitrite concentrations are given in Bal Krishna and Sathasivan [13]. Total chlorine residuals were measured by DPD colorimetric method using a HACH pocket colorimeter. Total chlorine measurement had an experimental error of ± 0.03 mg/L. DOC was measured using the Sievers 5310 C Laboratory TOC analyzer with an experimental error of $\pm 5\%$. A portable pH meter (HACH 40 d) was used to measure pH and the measurement error was ± 0.1 .

For the samples obtained from the SWDS, nitrite and TAN were measured using Flow Injection Analysis as detailed in APHA [15]. Nitrite was measured using the sulphanilamide method. Nitrite and ammonia for studied systems were measured using two different instruments but the measurement principle is the same. Therefore, the results are comparable. Total chlorine residuals were measured by DPD colorimetric method using a HACH pocket colorimeter. Detailed measurement methods are presented in Sathasivan et al. [2].

3. Results and discussion

3.1. General characteristics of bulk water samples collected from the three different chloraminated systems

Total chlorine residuals at the first chloramination point were 1.7, 3.7 and 2.5 mg/L in SWDS, GAWSS, and lab-scale system, respectively. Chloramine residuals dropped along the distribution system and the measured total chlorine residuals at the sampling points of the SWDS, GAWSS, GAWSS[#] (collected in winter) and lab-scale system were 0.85, 3.19, 1.73 and 1.76 mg/L, respectively (Table 1). Although total chlorine-to-TAN ratio was maintained close to 5:1 at chloramination point of each distribution system, the ratios were in the range of 2.3:1–4.5:1 at the sampling points. Comparatively higher nitrite concentrations (> 0.014 mg-N/L) were measured in the samples obtained from the lab-scale system and GAWSS[#] than in other two samples. This could be due to higher AOB population in the bulk water samples or due to the higher nitrifier activities in the biofilms and sediments of the lab-scale system and

GAWSS[#] when compared with the samples collected from GAWSS (during summer) and the SWDS. The water contact surface area (biofilm area) to bulk water volume is higher in the GAWSS (where water spend the majority of travel time in the pipelines) than in the SWDS (where water spend the majority of travel time in the service reservoir) demonstrating the bacterial activities from the biofilm and sediment could have played a major role. Additionally, water retention time is approximately double in the winter than in the summer in the GAWSS [3] which shows water age also could have impacted on bacterial activities, thus the higher concentration of nitrite. Similarly, bacterial activities of biofilm and sediments could be higher in the lab-scale system due to the higher biofilm area to bulk water volume ratio than in the SWDS.

A variation in bulk water temperatures between the studied distribution systems was noted. Water is supplied through above ground pipelines which are exposed to very high temperature (up to 40°C), especially in summer in the GAWSS. On the other hand, bulk water temperatures vary from 12–25°C in the SWDS. Lab-scale system is controlled environment where the temperature was maintained 18–23°C. Such variation in water temperatures greatly influences the stability of chloramine residual micro biologically and chemically [16,17]. Other chemical parameters such as pH, DOC were similar between the sampling points (Table 1).

3.2. Total chlorine and nitrogenous compounds profiles in the bulk water samples

Total chlorine decay behaviors in the bulk water samples collected from the studied distribution systems were different, especially after the onset of nitrification. Total chlorine residual gradually decreased whereas TAN and nitrite concentrations remained relatively stable until 10 d of incubation in the sample obtained from the SWDS (Fig. 1A). When chloramine residuals dropped below 0.55 mg/L and free ammonia concentration was 0.16 mg-N/L, TAN level rapidly decreased and subsequently, nitrite level increased showing the start of severe nitrification. Substantial acceleration in chloramine decay can be seen after the onset of nitrification (Fig. 1A). Similar behavior was observed in all mildly nitrifying samples collected from the SWDS when water temperature was between 12 and 20°C and initial total chlorine residual was about 1.7 mg/L [2].

Total chlorine residuals (3.19 mg/L) decreased slowly whereas nitrite and TAN residuals remained relatively stable until 33 d (Fig. 1B) when the GAWSS samples collected in summer (20–23°C) was monitored. Nitrification triggered when total chlorine residual and free ammonia concentrations were 1.06 mg/L and 0.44 mg-N/L (Fig. 1B), respectively. Similar to the samples collected in summer, the onset of nitrification was observed when total chlorine and free ammonia concentrations were 1.01 mg/L and 0.45 mg-N/L, respectively in the samples collected from the same system during the winter (Fig. 1C). Contrary to the SWDS sample, a rapid drop in chloramine residual did not occur after the onset of nitrification. It indicates that the onset of nitrification may not always notably accelerate chloramine decay, as it is commonly believed or as reported previously in one system [2].

Table 1
Water quality parameters measured in the bulk water samples at the time of samples collection

Parameters	Origin of samples			
	SWDS	GAWSS	GAWSS#	Lab-scale system
Total chlorine (mg/L)	0.85 ± 0.05	3.19 ± 0.03	1.73 ± 0.03	1.76 ± 0.03
TAN (mg/L)	0.29 ± 0.004	0.71 ± 0.01	0.76 ± 0.006	0.48 ± 0.006
Nitrite (mg-N/L)	0.005 ± 0.002	0.007 ± 0.002	0.014 ± 0.002	0.017 ± 0.002
Nitrate (mg-N/L)	0.10 ± 0.002	0.035 ± 0.002	Nm	0.050 ± 0.002
Total chlorine-to-TAN mass ratio	2.9	4.5	2.3	3.7
DOC (mg/L)	3.0 ± 0.2	3.1 ± 0.2	2.90 ± 0.1	3.0 ± 0.2
pH	8.0 ± 0.1	7.82 ± 0.1	Nm	7.84 ± 0.1
Temperature (°C)	18.2 ± 1.0	22.0 ± 1.0	17 ± 1.0	19.0 ± 1.0

Nm; not measured
GAWSS; Goldfields and Agricultural Water Supply System
SWDS; Sydney Water Distribution System
#Bulk water sample collected in winter

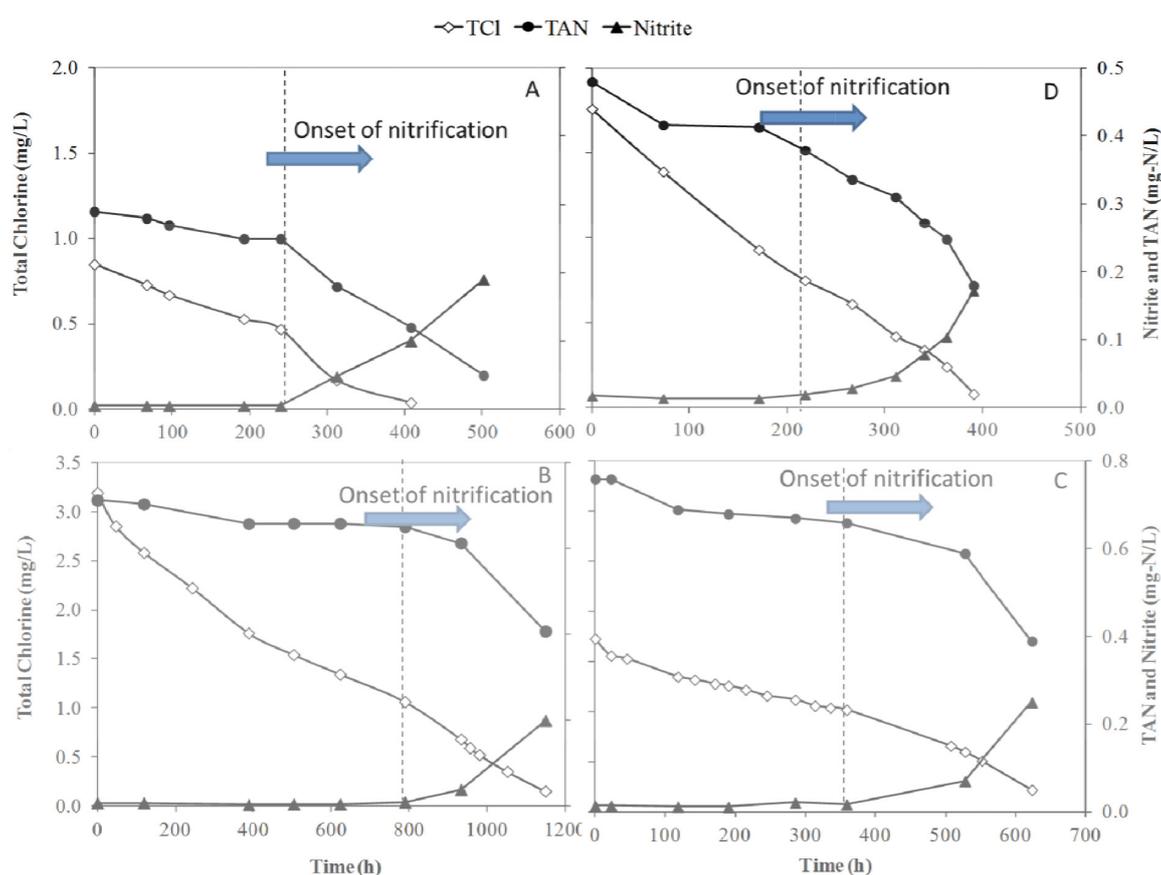


Fig. 1. Total chlorine, TAN and nitrite levels profiles, in the samples obtained from the three distribution systems having various initial chloramine residuals, 1A: SWDS, 1B: GAWSS, 1C: GAWSS# (samples collected in winter), 1D: Lab-scale System.

Although initial nitrite concentration (0.017 mg-N/L) was higher in the sample obtained from the lab-scale system than in other samples (Table 1), total chlorine and TAN concentrations remained relatively stable until 9 d of incubation (Fig. 1D). When total chlorine residual dropped below

0.7 mg/L and free ammonia residual was 0.23 mg-N/L, the onset of nitrification was noted. Similar to the SWDS sample, a rapid loss of chloramine residual was observed in the sample after the onset of nitrification (Fig. 1D). Such observation was noted when the experiment was repeatedly car-

ried out. The variation in chloramine decay profiles after the onset of nitrification between the samples obtained from different water distribution systems could be the result of several parameters such as rate of nitrite production, the release of soluble microbial products (SMP). Therefore, it is essential to separately quantify the microbial and chemical-induced chloramines decay before and after the onset of nitrification.

Moreover, samples used for the investigation contains different levels of total chlorine and TAN at the start-up of the experiment (Table 1). Both initial total chlorine and TAN concentrations impact on the growth of nitrifier hence on the onset of nitrification which is one of the reasons to see the occurrence of nitrification at different level of chlorine (Fig. 1). However, this study largely focused on determining the magnitude of chloramine decay as a result of the onset of nitrification in the bulk water samples. The difference in decay profiles could be the result of many other factors such as microbial species (types and population) and their activities, organic carbon (compositions and concentration), alkalinity which should be investigated in details.

3.3. Quantification of chemical and microbial-induced chloramine decay

The low F_m values (0.36–0.67) (Table 2) measured prior to the onset of nitrification demonstrate chemical reactions of chloramine with water constituents were the major cause of decaying chloramine residuals. Comparatively higher F_m values measured in the samples collected from the lab-scale system and SWDS before the onset of nitrification reveal the presence of higher microbial activities and potentially their higher population in those samples than the samples obtained from the GAWSS. After the onset of nitrification, significantly high F_m values were observed in all samples demonstrating the increased microbial activities and their role in the rapid loss of chloramine residuals. However, in this study F_m values are overestimated as the chemical decay coefficients (k_c) determined before the onset of nitrification was used to calculate the F_m . After the onset of nitrification, microbial metabolites such as nitrite and SMP are produced which increase the k_c values and reduce the actual F_m values.

Total decay coefficient (k_t) increased by 3.0–9.0 times in the bulk water samples (Table 2) after the onset of nitrification. Similar to F_m values, the high k_t values were noted in the samples collected from the SWDS and lab-scale system when compared with the samples from GAWSS. The increased k_t values were mainly the results of the higher microbial activities. The microbial decay coefficients (k_m) increased by 7.8–23.0 times after the onset of nitrification clearly proves the evidence of an increase of microbial activities in the samples whereas the k_c values were nearly the same between the samples (Table 2). Similar to F_m values, k_m values are overestimated as we have assigned the decay due to increased nitrite concentrations and potential release of SMP to the microbial decay. The k_m values measured in the samples obtained from the SWDS and lab-scale system were 2.8–3.5 times higher than in the samples obtained from GAWSS after the onset of nitrification. The high microbial decay could be the result of low chloramine residuals maintained in the distribution systems from where the samples were collected (Table 1) and the existence of high microbial activities (high k_m values when compared with GAWSS) prior to the occurrence of nitrification (Table 2). However, initial chloramine residuals were different (1.76 and 3.19 mg/L) in the samples collected from the GAWSS at different seasons but the k_m values were nearly the same (Table 2) indicating the initial low chloramine residuals only may not have facilitated for the higher microbial decay of chloramine.

3.4. Chlorine decay coefficients, k_t , k_m , and TAN drop rates in the bulk water samples

A good correlation ($R^2 = 0.953–0.979$) between TAN drop rates and chlorine decay coefficients (k_t and k_m) were noted after the onset of nitrification (Fig. 2). It shows the nitrification could be a major cause of the accelerated chloramine loss in the bulk water samples. The loss of TAN level usually increases the k_t due to the reaction between chloramine and nitrite. Our previous study [5] has demonstrated that SMP released by microbes after the onset of nitrification

Table 2
Details of microbial decay factor (F_m) related parameters

Parameters		SWDS	GAWSS	GAWSS#	Lab-scale system
k_t (h ⁻¹)	Before the onset of nitrification	0.0017 ± 0.0002	0.0015 ± 0.0002	0.0017 ± 0.001	0.0032 ± 0.0004
	After the onset of nitrification	0.015 ± 0.002	0.0053 ± 0.0012	0.0051 ± 0.001	0.013 ± 0.0015
k_c (h ⁻¹)		0.0011 ± 0.0002	0.0011 ± 0.0002	0.0012 ± 0.0005	0.0018 ± 0.0004
k_m (h ⁻¹)	Before the onset of nitrification	0.0006 ± 0.0004	0.0004 ± 0.0004	0.0005 ± 0.0015	0.0012 ± 0.0008
	After the onset of nitrification	0.0138 ± 0.0022	0.0042 ± 0.0014	0.0039 ± 0.0105	0.0112 ± 0.0019
F_m	Before the onset of nitrification	0.54	0.36	0.42	0.67
	After the onset of nitrification	11.5	3.8	3.25	6.23

Water sample collected in winter

GAWSS; Goldfields and Agricultural Water Supply System

SWDS; Sydney Water Distribution System

k_m ; microbial decay coefficient

k_c ; chemical decay coefficient

F_m ; Microbial decay factor (k_m/k_c)

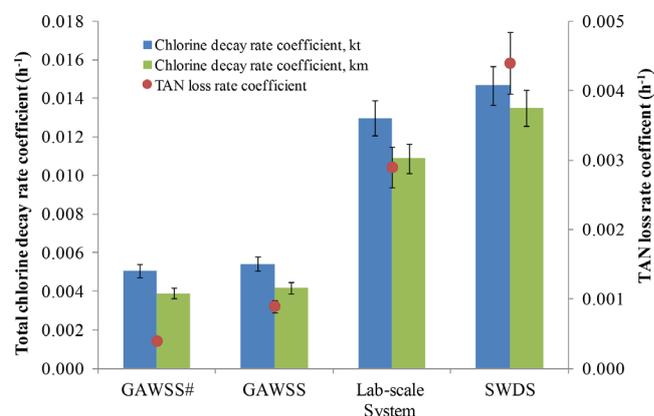


Fig. 2. Chlorine decay rate coefficients, k_t , k_m , and TAN loss rate coefficients after the onset of nitrification in the bulk water samples obtained from three different distribution systems.

catalytically accelerate chloramine decay. In addition, Herath et al. [18] showed heterotrophic bacteria may control chloramine decay in some cases. However, neither the role SMP nor heterotrophic bacteria have been quantified in this study.

In the chloraminated water distribution systems, nitrification rates depend on several factors such as total chlorine, free ammonia, pH, water temperature, dissolved organic carbon (types and concentrations) [7]. The pH and DOC concentrations were nearly the same between the samples and the same water temperature was adjusted in all samples during the study period. However, chlorine and free-ammonia concentrations were dissimilar between the samples. Free-ammonia concentrations were high in the GAWSS samples at the time of onset of nitrification. Moreover, previous studies [19,20] have shown that changes in overall bacterial communities including nitrifying bacteria with shifting chloramine residual and nitrification stage in the lab-scale system. As a result of varying chloramine residuals and other water quality parameters, the nitrifying bacterial species and their population might not be the same between the distribution systems. Moreover, different nitrifying bacterial species could have different means of oxidizing ammonia which could be one of the reasons for such observation.

Operating regime could be one of the reasons to observe such dissimilarity in chloramine decay behavior. In SWDS, operating temperature was between 12 and 25°C and concentration at the initial chloramination point was 1.7 mg/L [2]. In GAWSS system, water temperatures vary between 10 and 40°C and in the lab-scale system water sample was mildly nitrified and the temperature was between 18 and 20°C. Similarly, initial chloramine residuals maintained in the GAWSS and the lab-scale system were 3.7 mg/L and 2.5 mg/L, respectively. In the SWDS, GAWSS and lab-scale system, free ammonia concentrations were 0.12–0.20, 0.2–0.48 and 0.20–0.3 mg-N/L, respectively. These water quality parameters (chloramine residuals, free-ammonia residuals, and water temperatures) may select different microbial species and their population. Therefore, it is possible that the microbial

species survived in SWDS might not be present in the lab-scale system or GAWSS (collected in summer) sample at high chloramine residual as reported by [19].

4. Conclusions

The major conclusions made by the analysis are:

- The onset of nitrification was observed at a wide range of chloramine residuals (0.50–1.06 mg/L) in the samples collected from the different distribution systems.
- Higher chloramine decay coefficient was always observed after the onset of nitrification in each sample. However, between samples, the difference in decay rate was observed, i.e. in two samples total decay increased by about 3.0–3.5 times whereas in another two it increased by 4.0–9.0 times.
- A good correlation between TAN drop rates and chloramine decay rates were noted, but it is not completely proportional. The results indicated some other mechanisms could also be responsible for such decay.
- A different nitrite production rates between the samples could be due to the different strain of ammonia-oxidizing microbes and the number of such microbes present in the sample at the time of onset of nitrification.

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