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Modelling monochloramine decay and the impact of zinc in a simulated drinking water distribution system

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Abstract: Disinfection using monochloramine (NH_2Cl) is commonly performed for drinking water in distribution systems (DS) where there are long hydraulic retention times (HRT) involved in reaching customer taps. The use of monochloramine is due to its greater chemical stability and lower reactivity compared with other disinfectants, such a chlorine, enabling it to persist at effective concentrations in a DS over long distances and retention times. However, monochloramine undergoes decay in a DS, influenced by several factors including temperature, pH, HRT and pipework materials. Monochloramine decay can occur through chemical and microbiological processes, and this can be rapid through the latter process. Once established, microbiologically mediated monochloramine decay (MMMD) can be challenging to control as the microflora responsible for rapid decay can be present within the water as well as in biofilm around the pipework. Excessive decay can lead to the loss of an effective monochloramine residual leading to unacceptable risks from water-borne pathogens such as *Naegleri flowerii*. Potential procedures for managing rapid monochloramine decay in a DS include pH control and breakpoint chlorination.

Several studies have reported the potential use of metals that have anti-microbial activity including copper, silver and zinc for control of MMMD. These studies have predominantly been undertaken under short term-laboratory scale conditions. In the study reported here a pilot-scale distribution system (PDS) comprising a control stream (DS1) and a zinc treatment stream (DS2) was operated for ~ 4.5 years under field conditions, at a treatment plant that supplies chloraminated drinking water. After the establishment of MMMD in the first year of operation, the PDS was operated initially for laboratory and subsequently for field-based studies on the effect of zinc to mitigate this decay. In the field-based studies, several trials were conducted investigating the impact of zinc dosing at 3 and 6 mg Zn/L, with and without pH control. In assessing the potential impact of zinc on MMMD, mathematical models were generated to (1) describe the effects of HRT and temperature on decay as occurred in DS1, and (2) the effect of zinc dosed at 6 mg Zn/L with pH controlled at ~7.1, on mitigating decay in DS2. The model developed for DS1 (control) using data acquired over about 3.5 years was then applied to HRT and temperature data of DS1 and DS2 acquired during and after a zinc dosing trial (at 6 mg Zn/L, pH~7.1). No recovery of monochloramine residual was found during the period of zinc dosing. However, following discontinuation of zinc dosing, monochloramine residuals did recover in both DSs, though to a greater extent and for a longer period in the zinc treated DS. Mean ratios of the measured to model fitted NH_2Cl concentrations for DS1 and DS2 were 0.79 and 3.27 respectively, indicating that zinc dosing had led to monochloramine residual recovery. Based on this, two models were fitted to the monochloramine residual data, each to describe the recovery, maintenance and subsequent decline of monochloramine levels in DS2 during the period following zinc dosing. One model encompasses different rates for monochloramine residual recovery and subsequent decline with re-establishment of microbiologically mediated decay, while the other (Gaussian) is based on an assumption that the rate of recovery is equivalent to the rate of decline. Both model-fitted monochloramine (mg/L) data had high correlations with the measured monochloramine values ($r > 0.97$). Findings from this study indicate that these models might be applied for the testing of the impacts of antimicrobial metals on monochloramine decay and recovery under field-based trial conditions. These findings also indicate that targeted zinc treatment of sections of drinking water distribution systems impacted by MMMD has potential for reducing that impact and for the recovery of monochloramine residuals.

Keywords: *Monochloramine, Zinc, Drinking Water, Distribution.*

1. INTRODUCTION

One of the major challenges in operating a chloraminated drinking water distribution system is the prevention or minimization of monochloramine (NH_2Cl) loss that can occur through chemical and microbiological degradation processes. Free ammonia can be present in drinking waters of a distribution system due to the dosing of ammonia and chlorine for monochloramine formation or by the decay of monochloramine. Ammonia oxidizing bacteria (AOB) are then able to oxidize ammonia to nitrite and this in turn, can be further oxidized by nitrite oxidizing bacteria (NOB) to nitrate, referred to as the nitrification process (Bal Krishna et al. 2012). Factors that influence monochloramine decay in drinking water distribution systems (DWDS) include temperature, water age (or hydraulic retention time, HRT), piping materials and corrosion products and pH. Reported preventative and corrective strategies applied to control or avoid nitrification in chloraminated distribution systems include optimization of the chlorine-to-ammonia ratio, increasing chloramine residuals, addition of free chlorine to recombine this with excess free ammonia, periodic breakpoint chlorination, pH control, reduction of water retention time, water mixing in storage tanks, and removal of organic carbon (Bal Krishna and Sathasivan 2012).

Numerous studies have been conducted to investigate the factors influencing monochloramine decay and these have been performed at both laboratory and pilot plant scale (e.g. Gnos et al. 2013, Gomez-Alvarez et al. 2016). Several studies have been reported which investigated the potential use of heavy metals (e.g., silver, copper, and zinc) for inhibition of microbial activities responsible for rapid monochloramine decay. Bal Krishna et al. (2012) identified soluble microbial products which accelerated monochloramine decay and investigated the impact of protein denaturing agents including silver nitrate on monochloramine decay. They found that the dosing of silver at 0.05 – 0.15 mg/L, along with implementing protein denaturing pH and temperature conditions ceased rapid decay of monochloramine. Herath and Sathasivan (2020) found that chloramine decaying proteins (CDP) of 30-50 kDa, are responsible for catalytic acceleration of chloramine decay and reported that silver at 0.05 mg/L eliminated the impact of CDP. Sarker et al. (2018) investigated the effectiveness of using both zinc and copper. They found that 0.25 mg Zn/L inhibited nitrification and reduced the chloramine decay rate while 0.25 mg Zn/L with 0.2 mg Cu/L significantly controlled chloramine decay and inhibited nitrification completely. By comparison, Kulkarni et al. (2018), found that water samples collected from a pilot scale distribution system that exhibited high nitrification levels, and subsequently dosed with zinc under standardized laboratory conditions reduced microbiologically mediated monochloramine decay from 1.8 mg Zn/L. Although the effectiveness of zinc dosing has been investigated under standardized laboratory conditions, the effectiveness of zinc dosing under seasonally variable, field-based conditions requires investigation. Here we report the modelling of monochloramine residuals measured in a dual stream (control and zinc treatment) field-based pilot-scale distribution system (PDS), with established microbiologically mediated monochloramine decay. Zinc was selected for trials due to it being readily available, of relative low cost and being an essential element for human health.

2. MATERIALS AND METHODS

2.1 Pilot scale distribution system

Data were acquired from application of a PDS, previously described and detailed schematically by Kulkarni et al. (2020). The PDS was located at the Tailem Bend Water Treatment Plant (TBWTP), South Australia, (35.253º S, 139.456º E). The TBWTP is a conventional treatment plant (coagulation, flocculation, settling, filtration, monochloramine disinfection) and supplies drinking water over a network of ~280 km. The PDS comprised two parallel distribution streams, a control (DS1) stream and a zinc dosed (DS2) stream. Each stream comprised 900 m of 13mm polyethylene tubing with sampling points at 300m, 600m and 900m. The PDS was operated for about 4.5 years in total and in the first year MMMD was established. This was based on microbial decay factor (Fm), (Sathasivan et al. 2005) values for DS1 waters at 900m, which were >1. In the following 2 years, laboratory-based experiments were conducted to assess the effect of zinc on monochloramine decay (Kulkarni et al. 2018). Subsequently field trials were performed to further assess the impact of zinc dosing. In the original design of the pilot plant (Kulkarni et al. 2020), bulk water (water in which the DS 13 mm pipework was submerged) was used for moderating variations in water temperatures of the DS1 and DS2 waters inside the 13 mm pipework. However, following PDS maintenance after the third zinc dosing trial when potentially slow leaks were detected in both DSs, the use of bulk water for temperature moderation was discontinued. This was done to remove the risk of microbial contamination from the bulk water into the DS pipework.

2.2 On-line zinc dosing

A developed automated online dosing system previously described (Kulkarni 2020) was used for dosing zinc and pH control reagents. Water temperatures (T, ºC) and the pH of the inflow waters (TBWTP chloraminated

water) were measured. The pH of the inflow DS2 waters were adjusted to selected testing conditions as relative abundances of Zn^{2+} ions and zinc hydroxide ($Zn(OH)_2$ s) are pH and concentration dependent. The impact of the zinc speciation on monochloramine decay was unknown at the commencement of the study. Various zinc dosing trials were conducted including, Trial 1: 3 mg Zn/L without pH control and with a target HRT of ~8-10 days; Trial 2: 3 and 6 mg Zn/L dosing along with sodium hydroxide (NaOH) dosing intended for near optimum pH conditions (> 8) for monochloramine maintenance at HRTs ~ 8-10 days. Trial 3: 6 mg Zn/L with hydrochloric acid (HCl) dosing to lower the pH (~7.1) to promote elevated Zn^{2+} concentration and at HRT ~8-10 days. Zinc dosing was initially trialled at 3 mg/L, as that is the aesthetic limit for drinking water under the Australian Drinking Water Guidelines Version 4 (ADWG). However, as detected concentrations in DS2 were lower under the Trial 1 and initial Trial 2 conditions applied, potentially due to adsorption to biofilm, the dosing concentration was then increased to 6 mg/L.

2.3 Water Quality of TBWTP and PDS water samples

Data were acquired for water samples collected at the inflow to the PDS (from the TBWTP) and from three sampling points along each DS, i.e. 300m, 600m and 900m. These data included water temperature (°C), pH and monochloramine concentration. Monochloramine was analysed using DR 2800TM. Spectrophotometer and DPD Ferrous Titrimetric method (Kulkarni et al. 2018). Zinc concentrations were measured using Atomic Absorption Spectrophotometer as previously described (Kulkarni et al. 2018). The pH and temperature of waters were measured using a pH meter (WP-91, TPS Instruments) and digital temperature thermometer (TR-10, Elitech). MMMD was assessed by Fm values, as reported by Sathasivan et al. (2005). Fm is the ratio of the microbial decay coefficient (km) to the chemical decay coefficient (kc) of a first order decay function. This ratio was determined to differentiate between microbiologically mediated and chemical monochloramine decays in water samples (Sawade et al. 2016, Kulkarni et al. 2018, Kulkarni 2020).

2.4 Modelling of monochloramine residuals and the impact of zinc on residual decay.

HRTs at the three sampling points were determined from flow rates controlled by water pumps for each DS. For modelling of DS1 (control), data collected after the first year of operation were applied (excluding data during which MMMD was established). About 100 data sets (pH, temperature, HRTs and monochloramine concentration for each sampling point) were acquired. These data were grouped according to selected temperature (°C) ranges of (1) ~7 to 11, (n=7, $\bar{X}$ = 9.6), (2) ~12 to <15 (n=25, $\bar{X}$=13.1), (3) >15 to ~21 (n=28, $\bar{X}$=17.8), (4) >21 to ~25 (n=22, $\bar{X}$=23.2), (5) >25 to ~30 (n=8, $\bar{X}$=26.9), (6) >30 to <34 (n=5, $\bar{X}$= 31.7). For each group, mean HRTs (in days) and NH_2Cl concentrations (in mg/L) were determined. Model development for DS1 was based on the impacts of water temperature and HRT on residual monochloramine concentration. Generally, MMMD is positively correlated with these independent variables. The developed model was then applied to data acquired from DS2 (zinc treated) to assess the potential impacts of zinc.

Tablecurve © 2D Curve Fitting Hearne Software and Microsoft Excel were applied to generate models and for statistical analyses.

3. RESULTS AND DISCUSSION

3.1 Modelling monochloramine decay in the control distribution system (DS1)

As monochloramine dosing can be conducted at various concentrations, modelling of the monochloramine decay in the control DS was undertaken based on the percentage of the residual concentration measured in the DS to the dosed concentration. The purpose for modelling the % monochloramine decay in the control DS was to enable subsequent comparison to the zinc dosed (treatment) stream (DS2). The model of the % of monochloramine residual ((residual NH_2Cl /dosed NH_2Cl) *100) in DS1, is based on the means of HRT and T °C data that were grouped as previously detailed (Section 2.4), shown in Table 1.

Table 1. Percentages of mean NH_2Cl residuals (to dosed NH_2Cl concentrations) at the three mean HRTs for the various temperature groups.

Mean	Mean % NH_2Cl					
HRT\T°C	9.6°C	13.1°C	17.8°C	23.2°C	26.9°C	31.7°C
3.1 d	64.5	56.5	47.5	39.6	34.4	24.6
6.1 d	35.8	29.5	18.8	8.48	4.62	5.22
9.2 d	2.03	2.52	3.08	0.965	0.186	0.281

The linear function, y= a – bx was fitted to the $\%NH_2Cl$ and T°C data for each of the three mean HRTs (in days, d).

These are as follows,

for 3.1d, $\%NH_2Cl = 79.8 – 1.73*T$, ($R^2 = 0.9944$) (1)

for 6.1d, $\%NH_2Cl = 47.8 – 1.51*T$, ($R^2 = 0.9248$) (2)

for 9.2d, $\%NH_2Cl = 3.86 – 0.1155*T$, ($R^2 = 0.6511$) (3)

for the 'a' co-efficient,

$a = 98.38 – 3.377*(HRT)^{1.5}$, (4)

and for the 'b' co-efficient,

$Ln(b) = 0.54867 – 0.000277e^{(HRT)}$ (5)

From the above, the following relationship was generated,

$\%NH_2Cl = (98.38 – 3.377*(HRT)^{1.5}) – (e^x)*T$, where $x = 0.54867 – 0.000277e^{(HRT)}$ (6)

The above equation was applied using T°C and HRT input data, shown in Table 1, to generate model-fitted NH_2Cl values. These are compared to the group means of the measured NH_2Cl data, shown in Figure 1.

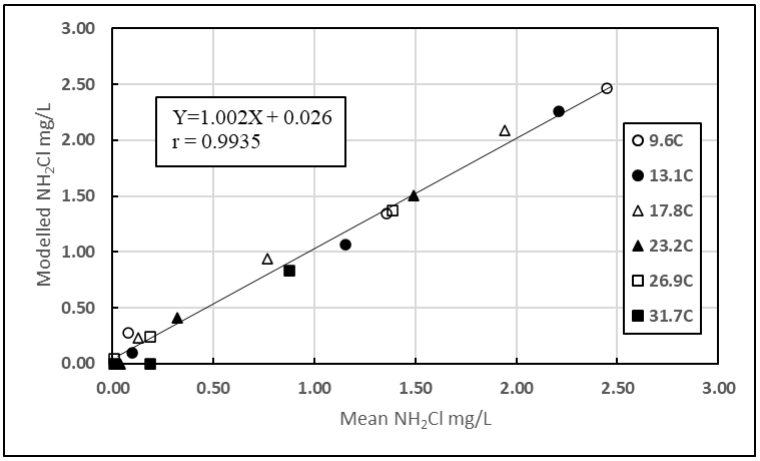

Figure 1. Correlation between model fitted NH_2Cl concentrations and the means of the measured NH_2Cl for the grouped temperatures.

3.2 Investigation of the impact of zinc on monochloramine decay.

During and after the application of zinc dosing at 3 or 6 mg/L in the first two trials conducted, there was no recovery of monochloramine residuals (<0.1 mg/L) at 900m of the DS2. During these trials, the monochloramine concentrations in DS1 at 900m were also either not detected or very low (<0.1 mg/L). These two trials were conducted during seasons when mean water temperatures in the two DSs were relatively high at ~18°C or more which then promoted MMMD. Also, in these two trials, the pH was either not adjusted or NaOH had been dosed with the aim of maintaining the pH at which monochloramine residuals are optimized. As there was no recovery of monochloramine at 900m (representing the point where long HRTs occur) in DS2 in trials 1 and 2, the pH of the zinc dosed (6 mg/L) water in Trial 3 was lowered (to ~7.1) with an objective of having zinc predominantly in its Zn^{2+} form. It was considered that in its free ion form that zinc would likely be more effective for inhibiting microbial consortia responsible for monochloramine decay and that this could lead to monochloramine recovery at long HRTs.

Trial 3 was also conducted in the winter period when water temperatures were lower than the previous trials which had been conducted in late summer and autumn seasons. It was further considered that these lower temperatures might also inhibit nitrifying microbial activities responsible for microbial monochloramine decay. However, in DS1 waters at 900 m during Trial 3 (mean pH 7.76), monochloramine levels were either not detected or at very low concentrations (Figure 2a). Also, during Trial 3, no recovery of monochloramine residual was found at 900m in DS2 (Figure 2b) and this likely due to the resultant low pH levels in the DS2 waters ($\bar{X}$ = pH 7.15) leading to loss of monochloramine through chemical decomposition. Vikesland et al.

(2001) reported the formation of dichloramine through monochloramine hydrolysis by a general acid catalysed monochloramine disproportionation reaction. Following completion of zinc dosing for 36 days, the operation of the PDS was continued, during which monochloramine recovery occurred predominantly in DS2 (Figure 2b), and to a lesser extent in DS1 (Figure 2a). Fm values were determined for the DS waters collected at the 900m sampling points. For DS1, Fm values were 0.6 on day 86 and 0.7 on day 128, while DS2 values were 0.001 and 0.6, respectively. These data indicate that MMMD was negligible in DS2 on day 86, but had re-established by day 128. By comparison, MMMD was evident in DS1 on both days 86 and 128.

For assessment of the extent of the impact of zinc, Eq. 6 was applied to HRT and T°C data for both DS1 and DS2 acquired during Trial 3 period of zinc dosing and afterwards. As the model equation was developed from long-term (~3.5 years) data acquisition, the impacts of temperature and HRT on NH_2Cl encapsulated a wide range of temperatures, as well as HRT variations. Modelled NH_2Cl data were acquired for each DS over the zinc dosing (A) and post zinc dosing periods (B). Mean ratios of measured NH_2Cl to model NH_2Cl data along with pH levels, are shown in Table 2.

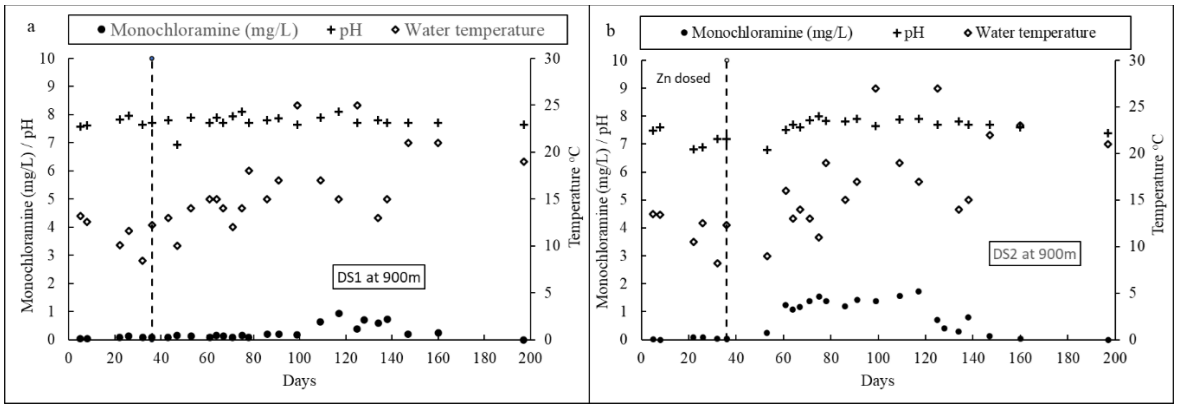

Figure 2: Residual monochloramine concentrations at 900m in (a) DS1 and (b) DS2 during and after Trial 3.

Table 2: Mean ratios of measured to model fitted NH_2Cl concentrations and pH levels, for DS1 and DS2.

DS	Trial 3 Period	n	Parameter	Sampling Point		
				300m	600m	900m
1, Control	A	6	R*	1.06	1.41	0.20
			pH	8.18	7.89	7.73
	B	20	R	1.16	1.78	0.79
			pH	8.26	8.09	7.76
2, Treated	A: Zn Dosed	5	R	0.45	0.24	0.11
			pH	7.07	7.18	7.19
	B: Post Zn Dosing	18	R	1.12	1.68	3.27
			pH	8.09	7.95	7.68

R*, ratio of measured to modelled NH_2Cl

During the period of zinc dosing (A) the measured NH_2Cl concentrations throughout DS2 were much lower than the modelled concentrations indicating the impact of low pH conditions caused chemical degradation of NH_2Cl. By comparison, the pH was higher in DS1 waters during Period A, and correspondingly, the measured to modelled NH_2Cl ratios were higher. During Period B, pH conditions in DS1 and DS2 waters were similar, but the measured NH_2Cl to model NH_2Cl ratio of DS2 was much greater (3.27) than DS1 (0.79) indicating that zinc treatment had impacted on the NH_2Cl concentration leading to the higher levels measured.

3.3 Modelling of the impact of zinc on monochloramine residual recovery in DS2

Based on zinc dosing in Trial 3 having had a subsequent beneficial impact on monochloramine residuals in DS2 for a discrete period of time, two models were fitted to data of the percentages of monochloramine residual (attributed to zinc dosing) to the original monochloramines doses. For this, the % NH_2Cl recovery (([DS2:NH_2Cl residual - DS1:NH_2Cl residual]/NH_2Cl dose)*100) data, abbreviated (%NH_2Cl_{rec}) were determined. The fitted models of these data versus days from discontinuation of zinc dosing are shown in figures 3a and 3b. The first model (Figure 3a) assumes the rate of monochloramine recovery is

different to the rate of subsequent decay with re-establishment of MMMD, while the second model assumes that these rates are equivalent. The models are as follows:

For Figure 3a, $\%NH_2Clrec=(a/(1+EXP(-(Days - b + c/2)/d)))*(1-1/(1+EXP(-(HRT-b-c/2)/e)))$ (7)

Where a = 29.32, the maximum modelled $\%NH_2CLrec$; b = 52.23, the mid-point of recovery range: c = 61.69, period of recovery; d = 2.57, rate of recovery; e = 5.92, rate of subsequent decline.

For Figure 3b, $\%NH_2Cl_{rec}= f + g*EXP(-0.5*(\left|\right. Days - h \left|\right. /j)^k)$, (8)

where f=0.273, the minimum $\%NH_2Cl_{rec}$; g = 29.1, the maximum level of $\%NH_2Cl_{rec}$; h = 52.6, mid-point of the zinc- effective range; Days = days since discontinuation of zinc dosing; j = 29.5, the 'zinc-effective' period (days) from the mid-point of the maximum 'zinc effective period (~ ≥18%) and k = 5.81, the rate of change from the maximum effective region of zinc.
The correlation co-efficient (r) values for the measured versus NH_2Cl_{rec} (mg/L) data are 0.9796 using Equation 7 and 0.9738 using Equation 8.

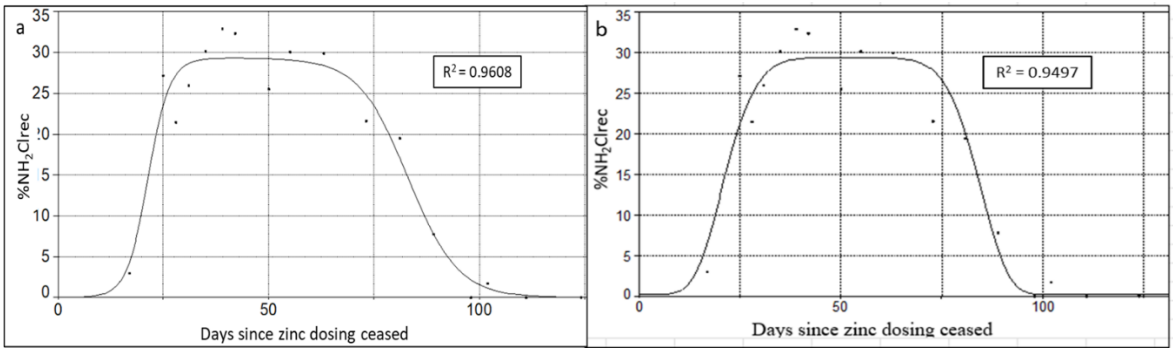

Figure 3. Comparison of measured and modelled $\%NH_2Clrec$ changes in DS2 at 900m (~ 9 days HRT) after Trial 3 zinc dosing using (a) Equation 7 and (b) Equation 8.

Both models described the features of the recovery of NH_2Cl residuals following discontinuation of zinc dosing when the pH is no longer lowered due to the use of HCl with zinc dosing, a period where NH_2Cl residuals are maintained and subsequently, a decline in the residual due to the re-establishment of MMMD. The findings from this study indicate that such models could provide insight into the impacts of anti-bacterial metals on monochloramine decay in field-based studies, where conditions such as temperature, HRT and pH are variable. It is suggested that such models have potential to be applied to other situations where periodic treatments are applied for monochloramine residual recovery and maintenance. In the case of any potential future application of anti-bacterial metals, such as zinc, to water distribution systems, compliance to regulated and/or guideline concentration limits would need to be ensured.

4. CONCLUSION

A field-based pilot plant study was conducted over an extended period (~4.5 years) to investigate the potential impact of zinc on MMMD in drinking water distribution with long HRTs. Following establishment of MMMD, several trials were conducted where zinc was dosed at either 3 or 6 mg Zn/L, with and without pH control. Monochloramine residual recovery at the 900m sampling point (HRT ~9 days) was not detected during and following dosing at 3 mg Zn/L, when the water pH was ambient or elevated using sodium hydroxide, and water temperatures were relatively high. In the third trial conducted, zinc dosing was tested at 6 mg Zn/L during a winter period when water temperatures were comparatively lower than previous trials, and with the pH lowered to maximize zinc presence in its free metal ion form. Monochloramine recovery was not found during the period of zinc dosing but was found subsequently, with greater recovery in the zinc treated than the control stream. A mathematical model was developed using data of the control stream acquired over the period following MMMD establishment (~3.5 years). This model relates the $\%NH_2Cl$ residual (to the dosed concentration) with plant water temperatures that ranged over the four seasons at the study site and variation in HRTs. The model was then applied to the control and treatment stream data acquired during and following Trial 3. For the 900m sampling point representing long HRTs of ~9 days, the mean ratio of the measured to modelled monochloramine concentration was much lower in the control (0.79) than the zinc dosed stream (3.27), a factor of ~4.1. This demonstrating the impact of zinc on recovery of monochloramine residuals. Two models, (1) with a different rate of monochloramine recovery to the rate of decline and (2) where the rate of recovery is equivalent to the rate of decline, were fitted to describe the recovery, maintenance and subsequent

decline of monochloramine concentrations, during the post-Trial 3 period. Although both models similarly described the post-Trial 3 data, it is likely that the first would more suitably describe the monochloramine dynamics in response to zinc treatment as trialled. The findings from this study indicate that zinc application, in its free ion (Zn^{2+}) form, has potential for the recovery of monochloramine residuals in sections of distribution systems where MMMD occurs. In the study conducted, dosing of zinc at 3 mg/L at pH ~ 7.1 was not investigated, and this is suggested for future investigation. Investigation by field-based pilot plant trials is also suggested to determine the impact of short-term dosing of higher concentrations of zinc (than used in this study) on MMMD and monochloramine recovery. For any dosing of zinc above regulatory or guideline limits (3 mg/L under the ADWG) into distributed drinking waters, the zinc dosed water would need to be applied as short-term flushing, followed by discarding of all of the dosed waters.

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