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Impact of zinc on biologically mediated monochloramine decay in waters from a field based pilot scale drinking water distribution system

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HIGHLIGHTS

- A field based novel design pilot-plant distribution system was designed and operated.
- Zinc below drinking water aesthetic limits lowered the monochloramine decay rate.
- Monochloramine decay rate and Fm values decreased as applied zinc doses increased.
- Efficiency of applied zinc doses decreased as initial microbial activity increased.
- Models of reductions in monochloramine decay rates with zinc addition were developed.

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ABSTRACT

Zinc is a heavy metal that is essential for human health at trace levels. Zinc also has antibacterial properties and these were investigated on microbiological mediated monochloramine decay (measured as a microbial decay factor, Fm) occurring in chloraminated drinking water. Water samples displaying rapid chloramine decay were obtained using a field based novel design pilot-plant distribution system (PDS), located at the Tailem Bend water treatment plant, South Australia. The PDS comprised 1 kL tanks each holding 900 m of polyethylene tubing with sampling points at 300 m intervals. Flow rates were applied to achieve hydraulic retention times (HRTs) that simulated those of the full-scale distribution system. Total (k_t) and microbiological mediated (k_m) decay rates were determined for chloraminated waters as these passed through the PDS. Highest Fm values (2.1 ± 0.7) were found for waters collected at the outflow of the PDS pipework, demonstrating increase in microbiological mediated monochloramine decay with increased HRT. Zinc (0.6–60 mg/L) added to water samples was found to lower k_t, k_m and consequently Fm, as its concentration was increased. Mathematical models (R^2 > 0.9 and T-test value > 0.8) were developed that describe reductions in monochloramine decay rates with the zinc addition. Reduction in k_t and Fm in response to zinc were found to have an inverse correlation with initial microbial activity in the chloraminated waters. Zinc concentration below drinking water aesthetic limits (< 3.0 mg/L, ADWG) was also found to lower the monochloramine decay rate, indicating potential benefit at such levels in chloraminated distribution systems.

Abbreviations: AOB, ammonia-oxidising bacteria; k_c, chemical decay coefficient; DBPs, disinfection by products; HRT, hydraulic retention time; k_m, microbial decay coefficient; X_{max}, maximum reduction value of k_t or Fm; Fm, microbiological decay factor; k_{PDS}, monochloramine decay rate within the pilot distribution system; NOB, nitrite-oxidising bacteria; PDS, pilot distribution systems; PET, polyethylene terephthalate; Fm_{Red}, reduction value of Fm; k_{Red}, reduction value of k_t; TBWTP, Tailem Bend drinking water treatment plant; k_t, total decay coefficient

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1. Introduction

Chemical disinfection is used in the supply of drinking water to inactivate or kill pathogens found in source waters, such as rivers and reservoirs. Chemical disinfection is applied with the aim to maintain residual concentrations in potable water distribution systems through to the point supply to consumers to ensure safe water provision. Chemical disinfectants include chlorine, ozone, chlorine dioxide and monochloramine (formed from chlorine and ammonia) [1]. Monochloramine is generally considered more stable than chlorine in the distribution system which leads to a lower formation of disinfection by products (DBPs) such as trihalomethanes (THMs) and haloacetic acids (HAAs) [2,3]. In contrast, monochloramine in drinking water can form nitrogenous DBPs such as N-nitrosodimethylamine (NDMA) [4]. Sakai et al. [5] reported that NDMA is a major contributor to overall lifetime cancer risk.

In Australia, monochloramine is used where drinking water supply is over long distances i.e. distribution systems have long hydraulic retention times (HRTs) due to its general high stability and effectiveness in providing protection from pathogenic micro-organisms such as the amoeba, *Naegleria fowleri*. Although the overall rate of chloramine decay by auto-decomposition is generally much lower compared with the rate of chlorine decay [6,7], monochloramine will nonetheless degrade over time. A number of chemical and biological factors influence monochloramine decay in drinking water distribution pipelines. These include nitrification [8,9], pH, temperature, Cl_2/NH_3-N_2 ratio [10], and reaction with organic substances present in treated waters [11,12]. Factors such as temperature, pH and initial chloramine concentration have consistent effects on auto-decomposition of monochloramine as the decay rate is known to increase with decreasing pH (from the optimum for this disinfectant), lower initial concentration of chloramine and increasing water temperature [13,14].

Nitrification has been reported to be a common operational problem associated with chloramination [9] caused by nitrifying bacteria where ammonia produced by the decomposition of chloramine, is subsequently oxidised to nitrite by ammonia-oxidising bacteria (AOB) and then to nitrate by nitrite-oxidising bacteria (NOB). Regan et al. [15] reported that *Nitrosomonas (Nm. oligotropha cluster)* and *Nitrospira* are the main AOB and NOB respectively, of the chloraminated distribution systems they studied. Cunliffe [16] reported that 64% of water samples collected from chloraminated distribution systems in South Australia contained nitrifying bacteria, and these being present in 20% of samples of waters with more than 5 mg/L monochloramine. From a field study in New South Wales, Australia, Fisher et al. [17] concluded that when chloramine residuals fall below 0.65 mg/L, there is a greater chance of nitrification occurring. In a study by Fleming et al. [18] incorporating a pilot scale distribution system, concentrations of free ammonia and total chlorine were linked to the growth or inactivation of AOB and consequently associated with the nitrification levels.

In a bench-scale simulated distribution system, Pintar and Slawson [10] reported the presence of AOB at various temperatures (12 and 22 °C) for waters with chloramine residuals ranging between 0.05 and 0.6 mg/L. In laboratory scale reactors, Krishna and Sathasivan [19] reported that nitrification levels (measured as nitrite, nitrate and total ammonical nitrogen) affect chemical decay rates with the decay coefficient higher in highly nitrified waters. Also using laboratory scale reactors, Zhang et al. [20] reported that AOB abundance was higher in reactors that had high concentration of natural organic matter. Frias et al. [11] reported an increase in the number of circulating bacteria after the addition of organic carbon to a pilot scale chloraminated distribution system.

Nitrification has been reported to be inhibited by metals such as copper, lead and zinc in wastewater treatment processes [21,22]. Nitrifying bacteria (ammonia and nitrite oxidising bacteria) present in wastewater samples from eight different industries in Sweden, including the printing industry and industrial laundry, have been found to be inhibited by zinc at concentrations ranging between 0.003 and 1 mg/L [21]. Depending on its concentration and contact time with AOB, zinc affects the rate of nitrite production [23] by AOB, by binding onto the active site of ammonia monooxygenase of AOB. Although studies have investigated the effects of zinc on nitrification during wastewater treatment processes as exemplified above, there is a lack of studies on the effects of zinc on drinking water quality in relation to monochloramine decay in water distribution systems. Under the Australian Drinking Water Guidelines [24] the aesthetic limit for zinc in drinking water is 3 mg/L and there is currently no limit set for zinc based on human health considerations. Similarly, the US EPA [25], the WHO [26] and the EU [27] do not have listed any limits on zinc concentration in drinking water based on human health considerations.

In drinking water distribution systems, problems caused by low chloramine residuals include microbial regrowth [28], nitrification [29] with reduction in alkalinity levels [30] and remedial action (e.g., dosing of free chlorine) is required to avoid complete loss of monochloramine residual [31]. Monochloramine is applied for disinfection of drinking water that is supplied over long distances. Rapid biologically mediated monochloramine decay in drinking water distribution systems occurs generally in systems with long HRT, and this remains a significant problem for the water industry. In this paper, we report the design and operation of a novel field based PDS operated at flows that simulated long HRTs > 5 days, to promote rapid monochloramine decay. Using this PDS the effects of low to high concentrations of zinc (0.6–60 mg/L) on rapid monochloramine decay, was studied. Mathematical models were developed to describe the reductions in monochloramine decay rates resulting from the zinc doses applied. In this study, the microbiological decay factor (Fm) [32], was used to assess the impacts of zinc on microbiological (or biologically) mediated chloramine decay.

2. Materials and methods

2.1. Pilot distribution system (PDS)

A schematic diagram of the PDS is shown in Fig. 1. The PDS was set up at the Tailem Bend Drinking Water Treatment Plant (TBWTP), Tailem Bend, South Australia (35.274° S, 139.459° E). The water treatment process (capacity of 28 ML/d) comprises coagulation & flocculation, tube settler clarification, dual media rapid gravity filters, and disinfection by UV (low pressure mercury lamps) and monochloramine. Powdered activated carbon is periodically used and the treated water is fluoridated prior to distribution.

The PDS consists of two identical cylindrical polyethylene tanks of 1 m diameter and 1.8 m height (~1 kL capacity each). Each tank consists of 3 bundles of polyethylene PE tubing (13 mm × 300 m each) connected in series with supporting connections and sampling points (at the inflow, 300 m, 600 m and 900 m). The pipework was designed to provide controllable hydraulic retention times that simulated long hydraulic retention times of full scale networks. The polyethylene tubing size (13 mm internal diameter) used in the PDS provided a comparatively high surface area to bulk water volume, with the ratio of circumference to cross sectional area being ~0.31 compared to full-scale distribution systems (e.g. for a 100 mm pipe, the ratio is ~0.04). This was designed to promote biofilm growth and microflora proliferation in the PDS. These tubing bundles were held immersed in treated water sourced from TBWTP that continually flowed through the tanks externally to the tubing in order to stabilise the temperatures. Without this, the water inside of the pipework would experience rapid changes in daily temperatures. For the study reported here, water samples from one of two identical tanks of the PDS were used.

The PDS is supplied with three optional water sources, raw water (sourced from the River Murray), filtered non-chloraminated water and fully treated water from the TBWTP. At the commencement of its operation, the PDS was initially supplied with a mix of raw (80%) and