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Water Impact

RWDSs are integral to achieving water sustainability. The unique chemistry and microbial characteristics of reclaimed waters, require improved understanding in order to manage RWDSs in a manner that achieves objectives at the point of use. This laboratory simulation discovered that pretreatments, distribution system design and operation can profoundly affect sediment accumulation, which in turn impact disinfectant depletion, microbial growth and water quality.

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Abstract

Changing chemistry and microbiology of reclaimed water as it is conveyed via reclaimed water distribution systems (RWDSs) can influence water quality at the point of use. Two simulated RWDSs comprised of 0.32-cm tubes ("Tube" RWDSs) and 10-cm diameter pipes ("Pipe" RWDSs) were designed to investigate how extremes in water age (32-min versus 5-d) and surface area to volume ratio (6.25 cm^{-1} versus 0.20 cm^{-1}) affect the potential for sediment accumulation, disinfectant decay, and chemical and microbial water quality under controlled treatment conditions. Effluent from a conventional municipal wastewater treatment plant was breakpoint chlorinated and treated with and without biological filtration, followed by three disinfectant conditions: no residual, 4 mg/L chlorine or 4 mg/L chloramine. After three years of operation, accumulated sediment occupied 0.32-3.2% of the total volume of the first Pipe

segment. The unfiltered chlorine condition had an order of magnitude less influent turbidity and sediment accumulation than conditions with no residual or chloramine, resulting in lower levels of biological activity and less depletion of disinfectants and dissolved oxygen. The sediment in Pipes receiving chloraminated water was highly biologically active and created a high disinfectant demand due to nitrification. In contrast, the Tub RWDSs did not accumulate sediment. The Tube rigs with chloramine and no residuals had heavy bio-foulant growth, which resulted in rapid depletion of chloramine. Biofilm was the main source of disinfectant demand in the chlorine Tubes. An improved holistic understanding of RWDSs chemistry, biology and operation will help achieve desired water qualities at the point of use.

1. Introduction

Population growth, climate change, and increased water scarcity necessitate sustainable water management that includes reuse of moderate to highly treated wastewater effluent.[1] For instance, the Inland Empire Utilities Agency in San Bernardino County currently reuses 50% of its wastewater and is targeting 80% reuse by 2025.[2] The level of water reuse treatment should be tailored for its intended use, ranging from conventional secondary wastewater treatment plant (WWTP) effluent with disinfection that delivers beneficial nutrients for crop irrigation, to technologies that can produce water far exceeding potable water standards.[1]

More than 50% of reuse in the United States is directed towards non-potable applications, such as agricultural, urban, and industrial reuse.[3] Such applications do not generally target extensive removal of suspended solids or pathogens beyond conventional disinfection. Most state regulations for distributing reclaimed water allow up to 5 mg/L total suspended solids, which roughly translates to 5-10 Nephelometric Turbidity Units (NTU),[4] whereas drinking water turbidity is often rigidly controlled to less than 0.03 NTU.[1] This creates a much greater

potential for sediment accumulation in reclaimed water distribution systems (RWDSs) versus drinking water distribution systems (DWDSs).

According to a WateReuse Research Foundation survey of US reclaimed water system operators, even though they do not usually expect non-turbid or aesthetically pleasing water, 12% cite "biofilm growth," 5% report "sediment accumulation," and 25% note frequent problems with "clogging of sprinkler heads" as issues of concern in reclaimed water.[5] The effective removal of particulates is a primary goal of drinking water treatment using conventional sedimentation and filtration, but even so, recent research demonstrated that accumulated loose deposits in highly treated water distributed through a polyvinyl chloride (PVC) pipe network in the Netherlands served as reservoirs for microorganisms, including opportunistic pathogens. [6,7] A nation-wide survey of drinking water storage tanks in the U.S, also revealed high sediment accumulation and growth of opportunistic pathogens with sediment.[8] All of this work suggests that stagnant or low flow portions of RWDSs are likely highly susceptible to sediment accumulation and microbial growth.

Understanding of key processes controlling the chemistry and microbiology of potable water distribution to ensure high quality water at the point-of-use has advanced substantially in recent decades. A knowledge base has been established about relative rates of decay for different disinfectants,[9,10] importance of nutrients,[11,12,13] and formation of distinct redox zones as a function of water age in DWDSs.[14,15] In recent years, understanding of water quality problems in DWDSs has been extended to a few studies of RWDSs, where impacts of high levels of particulates, organic matter, nutrients, and bacteria on the final water quality of distributed water are likely to be magnified.[16-19], Intermittent flow is another common characteristic of RWDSs, allowing for sedimentation of particulates and causing depletion of

secondary disinfectant residuals during stagnation events,[5,17] potentially leaving systems vulnerable to water quality degradation.[21,22]

There are currently no specific federal regulations controlling water reuse or distribution. Given the wide variation of influent water quality, treatment processes, water use patterns and intended end uses comparing DWDSs and RWDSs, improved fundamental understanding and management practices are needed. Here we employ two types of controlled, simulated RWDSs to better understand the role of sediments and biofilms in controlling water chemistry and overall microbial regrowth potential. The study design, included two of the most common treatments in reclaimed water systems, biologically activated carbon (BAC)-filtration and disinfection (i.e., free chlorine or chloramine) and simulated representative reclaimed water distribution system hydraulic regimes to facilitate comparison to on-going comparison to full-scale RWDSs. The specific objectives were to determine: 1) how different treatments, including BAC-filtration and disinfection, affect sediment composition and accumulation; 2) the role of different simulated RWDS design; and 3) the interplay between treatments and design that affect chemical redox zones and levels of secondary disinfectant residuals levels.

2. Materials and Methods

2.1 Simulated RWDS Influents

Final wastewater effluent from a conventional municipal WWTP was collected twice a week and stored at 4 °C before being subjected to various treatment scenarios. Half of the water was treated using BAC filters for 30 hours, which typically removed 25% - 33.3% of

the total organic carbon (TOC), with the other half remaining unfiltered (Table 1). The water was then breakpoint chlorinated to achieve a detectable chlorine residual of < 0.2 mg/L, to remove the chlorine demand and destroy ammonia in the feed water. The required breakpoint dose was 2.85 ± 1.87 mg/L for unfiltered water and 1.16 ± 0.06 mg/L for the BAC-filtered water. Three secondary disinfectant conditions were created for each water: no additional disinfectant, free chlorine at 4 mg/L as Cl_2, and chloramine at 4 mg/L as Cl_2 and a 4:1 weight ratio of final chlorine to ammonia to create monochloramine. The three secondary disinfectant conditions (chlorine, chloramine and no residual), both with and without prior BAC-filtration, resulted in six distinct influent water conditions (Table 1).

In order to achieve the goal of a stable disinfectant residual for the unfiltered 4 mg/L free chlorine condition, a two-step superchlorination process was used to remove the slow chlorine demand form the high level of organic matter in the unfiltered raw water. Eight milligrams per liter of Cl_2 was dosed the day before the water was used to prepare the feed and provide extended chlorine contact time to destroy chlorine demand. Right before using the water, additional chlorine was added to achieve 4 mg/L as Cl_2 in the reservoir. While it is recognized that the unfiltered chlorine condition experienced a much higher total chlorine contact time than the other disinfected conditions, this was necessary to maintain the starting residuals in all disinfected reservoirs at 4 mg/L as Cl_2.and have a consistent baseline to assess decay in the pipes across different conditions. Each influent reservoir was stored at 4 °C to minimize microbial activity as it was pumped into the simulated RWDSs. All reservoirs were changed every 30 hours.

The water age, or the time to travel from treatment plant to point of use, can vary from 0 to more than 24 days in real water systems, having a strong influence water quality at the

point of use.[23] Water pipes can also have inner diameters ranging from a few millimeters in buildings up to a few meters in water mains,[24] while flow can range from completely stagnant to turbulent. Two simulated RWDS rig designs – the "Pipe" and "Tube", were employed to represent extremes in terms of water age, flow patterns, surface area to volume (SAV) ratio, and likelihood of sediment accumulation (Figure 1).

The wide Pipe RWDSs were designed to achieve a realistic simulation of both water age and SAV ratio, albeit at an unrealistically low flow velocity. The complementary Tube RWDSs were designed to test a narrow pipe diameter that prevented sediment from accumulating and a higher internal flow velocity, but with an extremely low water age and high surface area to volume ratio. The Pipe rigs were comprised of three 10-cm (4-in.) diameter PVC pipe segments connected in series with short PVC tubing in between (Figure 1a). PVC was selected as a least reactive pipe material commonly used in full-scale distribution systems, relative to metallic, concrete and other materials,[25] in order to minimize the potentially important effect of pipe corrosion and dissolution for purposes of this work. The calculated water age (volume/flow rate) of the three pipe segments corresponded to 1 d, 2.5 d, and 5 d. The narrow Tube simulated RWDS rigs consisted of six 15.24-meter (50-ft) 0.32-cm (1/8-in.) inner diameter PVC tubing with a calculated water age of 32-min (Figure 1b). The SAV ratio was 6.25 cm^{-1} and 0.20 cm^{-1} for the tube and pipe RWDS's, respectively. Three disinfectant conditions (no disinfectant, chlorine, chloramine) applied to unfiltered water were operated in duplicate for the Tube rigs. The flow rate for the Pipe and Tube rigs was identical at 4.2 ml/min.

Prior to the experiment described herein, all Pipe rigs had been operating under the targeted influent conditions for 39 months (June 2015 – May 2017) to allow a mature biofilm

to establish. There was a one-time repair and replacement of a leaking first segment of the unfiltered chloramine Pipe rig in Sep 2016. The repaired Pipe remained in continuous operation over the subsequent 24 months. The Tube rigs were first operated from April 2017 – August 2017 (5 months), left stagnant for eight months to simulate an extreme non-use event, and then reconditioned April 2018 – August 2018 (5 months). To ensure a valid head-to-head comparison between the two rigs, all rigs were operated under the same ambient temperature (~22 °C) and influent water conditions from April – August 2018. The same batch of effluent water was used to prepare the influent reservoirs during the sampling events to eliminate any background variation in influent water quality.

2.2 Tracer Study

A tracer study was conducted to define the hydraulic flow patterns of the rigs. Potassium chloride (KCl) was injected to the influent of one representative Pipe (at 50,000 mg/L KCl) and Tube rigs for each of the three disinfectant conditions (at 30,000 mg/L), with all effluent collected in aliquots until the potassium level returned to approximately the initial background levels. A clean rig without any sediment was tested as a control.

2.3 Sample Collection and Water Quality Analysis

The Pipe rigs were sampled both along the horizontal flow direction at 0%, 20%, 50% and 100% of the pipe length and along the vertical pipe profile at 0% (top), 50% (middle), and 100% (bottom) of the depth of the 10-cm pipes to explore variations in water chemistry in proximity to the sediment (Figure 1). To reduce effects of disturbances on sample results, measurements were made in reverse order from highest to lowest water age and from top to bottom. Routine samples of Tube rigs were from the effluent, whereas end of study profile

samples were collected by cutting the Tubes at sequential 10-foot (3.05 meter) intervals and collecting the effluent at 40, 30, 20 and 10 feet.

After the final sampling event, all water and accumulated sediment from the first pipe segment were transferred into separate buckets. The water and sediment mixture was settled and carefully decanted to avoid disturbance of settled sediment. The remaining 500 mL of water and sediment mixture was poured into a graduated cylinder. After allowing for 30 min of settling, the supernatant bulk water was carefully pipetted out without disturbing the settled sediment and the final settled volume was determined. The resulting sediment was analyzed for volatile solids (VS) and total solids (TS) according to EPA Method 1684.[26] All containers employed in sediment harvesting and quantification were sterilized either by ethanol or autoclave.

Water chemistry was tracked in all simulated RWDS rigs, including chlorine and chloramine residuals (HACH DR 2700 Spectrophotometer Method 8021), DO (Orion Star A 326 DO meter), disinfection by-products (DBPs) (EPA Method 502.2), pH (Oakton pH 110), ammonia (Hach Method 8155), nitrite and nitrate (Dionex® DX-120 Ion Chromatograph), and TOC (Sievers 5310C Laboratory TOC Analyzer).

Microbial characterization included nitrifiers (HACH N-BART™ Test), denitrifiers (HACH DN-BART™ Test), and total cell counts using a BD Acccuri C6® flow cytometer and previously reported methods.[27] Heterotrophic plate counts (HPCs) were used as an indicator of general microbial activity in both bulk water and in collected sediment.[28] Assimilable organic carbon (AOC) was measured using the method described by Hammes and Egli.[29] One milliliter of wastewater effluent without any on-site laboratory treatment was used as the indigenous bacterial consortium inoculum to seed 50 mL for AOC analysis. The

bacterial growth stage was carried out at 35°C for 5 days. The net increase in biomass over the 5-day incubation was measured by flow cytometry (BD Accuri C6®) and converted to equivalent AOC. Soluble AOC was measured after filtering the water through a 0.22 μm filter (Millipore® Millex® sterile syringe filters, Durapore® PVDF membrane). Total AOC was measured by replacing filtration with pasteurization at 70°C for 30 min, as previously described.[30]

2.4 Disinfectant Demand Characterization

The relative contribution of disinfectant demand from bulk water, biofilm, and sediment in the filtered and unfiltered chlorine and chloramine Pipes was measured in a flask test (Figure S1). Flasks (250 mL) included bulk water as the control, bulk water with 10 cm^2 swab of biofilm from a chlorine/chloramine Pipe wall added (the cotton swabs were verified not to incur additional chlorine demand in a side test), and bulk water with 10 mL sediment collected from the chlorine/chloramine Pipe added. All flasks were initiated with 4 mg/L of either free chlorine or chloramine and were monitored for up to 5 days for free/total chlorine residuals to establish decay curves for each condition.

2.5 Data analysis and statistics

Pearson's correlation test between influent turbidity and accumulated sediment volume was conducted with the ggpubr package in R and plots were produced with the ggplot2 package in R. First order decay model with a fixed initial disinfectant level of 4 mg/L was used to fit the disinfectant decay data in the flask decay study.

3. Results and Discussion

3.1 Characteristics of sediment in the Pipe Rigs and Tube Rigs

3.1.1 Sediment accumulation in the Pipe Rigs

After three years of operation, substantial quantities of sediment accumulated in the bottom of the first pipe segment of the Pipe RWDSs. The accumulated sediment appeared loose, flocculent, and dark (Figure 2) and was obviously distinct from the thin, slimy biofilm layers distributed more uniformly around the inner circumference of the pipe surface. The total sediment volume in each pipe reactor ranged from 19 to 190 mL (Table 1). The greatest sediment volume was observed in the condition with no filtration or disinfectant residual, occupying 3.2% of the 6 L total volume of this segment, whereas sediment in the filtered chlorine condition occupied just 0.32% of the total volume. The VS/TS ratio of the sediment was independent of whether or not the water had been biologically filtered, but could be ranked according to disinfectant condition: chloramine (82.9% and 79.1%) > no disinfectant residual (76.9% and 76.4%) > chlorine (48.1% and 50.9%) (Table 1). The high VS/TS ratio in the chloramine and no disinfectant residual conditions indicates a higher fraction of viable biomass,[31] relative to the 60% VS/TS ratio reported for DWDS sediment[32] or the corresponding condition with free chlorine reported herein. Suspended solids in secondary reclaimed water effluent are known to contain high levels of organic particulates that produce biologically-active sediments in lakes.[33,34] Such effects were observed for all RWDS conditions without free chlorine tested herein.

The two most likely sources of sediment accumulation in the Pipes were physical settling of suspended influent particulate matter or sloughed biofilm. The representative measured influent turbidity varied from 0.78 – 12.6 NTU (Table 1), where expectations that the BAC-

filtered influent would consistently have lower turbidity than the corresponding disinfectant conditions without filtration was confirmed. However, the high levels of free chlorine residual destroyed turbidity, presumably via direct reactions with natural organic matter and particulate organics, producing influent with low turbidity of 0.78 – 1.20 NTU in water with or without filtration. There was a strong positive correlation between influent turbidity and sediment mass and total sediment volume, supporting the notion that the sediment was largely derived from settling of influent particulates (Pearson's correlation, r = 0.91 and 0.89, Figure 3). The accumulation of sediment in the filtered chloramine Pipe was above the trend line, suggesting sloughing of biofilm might have been a more substantial contributor for this condition.

3.1.2 Bio-foulant accumulation in the Tube rigs

While sediment deposits were not observed at the bottom of the Tube rigs at the end of operation, "bio-foulant", which herein refers to symmetrical accumulation of biological and non-biological buildup on the Tube walls, was prevalent and varied among the disinfectant conditions (Figure 4). The bio-foulant thickness was very thin in the chlorine condition, consistent with observations that this condition never clogged at any point of the study. In contrast, the chloramine and no disinfectant residual conditions clogged repeatedly after two months of operation and had to be cleared by brief (10 seconds) reversal of the pump flow direction on a weekly basis.[35,36] The bio-foulant in the chlorine Tubes was also lighter in color than in the chloramine or no residual condition, possibly due to bleaching.[37] The trends in bio-foulant accumulation in the Tube rigs due to disinfectant type was similar to what was observed for sediment accumulation in the Pipe rigs. However, the design of the Tubes rigs

achieved prevention of deposit accumulation at the downward orientation as observed in the Pipe rigs.

3.2 Assessing the effects of rig design, sediment, and biofilm on tracer curves.

3.2.1 Pipe RWDS

The observed tracer curves varied depending on the accumulation of sediment and biofilm in the Pipe rigs. The tracer curve for a new Pipe section without any accumulated sediment and biofilm peaked at a residence time of 20 hours, with a mean tracer residence time of 33 hours. This was in the range of the calculated 26-hour hydraulic residence time for a plug-flow reactor (PFR). In contrast, the tracer curve from the first pipe segment with accumulated sediment and biofilm resulted in a curve more characteristic of a continuously-stirred tank reactor than a PFR (**Error! Reference source not found.**5). In the Pipe rigs with sediment accumulations (i.e., the filtered chloramine Pipe), the tracer peaked at 14 hours instead of 20 hours for a clean pipe, but it had a mean tracer residence time of 73.3 hours.

Clearly, the KCl tracer, which is often considered conservative and relatively non-reactive in DWDS applications,[38] was significantly retarded in the established Pipe RWDSs, especially when considering that the accumulated sediment was expected to create dead space that caused the tracer to elute more quickly than in the control pipe without sediment and biofilm accumulation. After repeating the tracer study several times, it was concluded that the KCl was not acting as an idealized conservative tracer under the biologically active conditions of the Pipe rigs, but was probably being actively accumulated in the abundant biomass, e.g., via the sodium-potassium pump mechanism.[39] Alternatively, biofilm has sorptive ion exchange properties for potassium, which could also cause retardation of the tracer elution from the reactors ions as a way to obtain nutrients for embedded cells in fixed biofilm.[40, 41,42]

3.2.2 Tube RWDS

KCl tracer studies confirmed a strong PFR flow characteristic for a new Tube rig or an established rig with relatively little bio-foulant (chlorinated reclaimed water), with peaks eluting near the calculated hydraulic residence time (Figure 6). However, in the corresponding rigs with visibly thick bio-foulant levels, the tracer took nearly twice as long to elute (Figure 4), consistent with the same counterintuitive result obtained in the Pipe RWDS rigs. This further supports the conclusion that KCl transport was retarded by the bio-foulant.

3.3 Sediment creates distinct redox zones in simulated RWDS

3.3.1 Disinfectant decay trends

The disinfectant residuals in the simulated RWDS rigs differed from what has been previously observed in DWDSs. As water passes through a DWDS, the level of disinfectant residual decays at a characteristic rate.[23] Here, both chlorine and chloramine residuals were completely depleted after the 5-day calculated hydraulic residence time for all conditions (Figure 7a). Contrary to expectations based on experiences in full-scale and some smaller-scale DWDSs,[43] chlorine residual was observed to be more persistent than chloramine residual in this simulated RWDS design. For example, ~1 mg/L chlorine residual still remained after 1 day for both the unfiltered and BAC-filtered waters in this testing at 22°C, whereas chloramine was between non-detectable to 0.06 mg/L.

A distinct disinfectant decay gradient along the vertical axis of the Pipes was observed within the first pipe segment, where nearly all of the disinfectant decay occurred. Specifically,

comparing sample aliquots collected from the top versus bottom of the Pipe rigs at the point of entry (P0), 2.61 mg/L of chloramine residual and 1.90 mg/L chlorine residual were lost in the unfiltered influent conditions as the sample approached the sediment at the bottom of the Pipe rigs (Figure 7b). The corresponding BAC-filtered conditions indicated similar disinfectant losses from top to bottom with 1.31 mg/L and 2.06 mg/L loss for chorine and chloramine, respectively. Sediment accumulation in the chloramine Pipes, which also had a much higher mass and VS/TS ratio than the corresponding sediment in chlorine Pipes, likely contributed to this faster rate of chloramine decay for both BAC-filtered and unfiltered water

3.3.2 DO consumption in the Pipe RWDS

DO is an important water quality parameter that is often used to track the extent of cellular respiration during transport and the likelihood of odors in water at the point of use.[44,45,46, 47] Injection of DO in reclaimed water distribution pipes can also improve the microbial quality of the delivered water.[48] In agriculture, low DO in the irrigation water can contribute to suboptimal crop growth, low yields, and plant diseases.[49] In the present study, DO generally decreased as water flowed horizontally through the biologically active Pipe rigs (Figure 8a), with a few slight exceptions where DO increased because the system was not designed to be completely impermeable to gases. Differences in DO losses were notable, with 3.22 or 5.41 mg/L DO lost for the BAC-filtered and unfiltered chloramine conditions, respectively, while only 0.38 – 1.14 mg/L was lost in the corresponding free chlorine conditions after 5 days.

As was the case for the disinfectant, DO decreased markedly, moving vertically from the top of the Pipe towards the sediment in the first pipe segment for both the chloramine and no residual conditions (Figure 8b). However, this trend was not observed in the chlorine

condition, presumably due to the much reduced accumulation of sediment and less biological activity. The DO drop from the top to the bottom of the first pipe segment ranged from 3.01 – 5.89 mg/L among the unfiltered or filtered influent conditions with chloramine or no disinfectant residual, compared to a drop of just 0.89 – 1.81 mg/L in the corresponding conditions with free chlorine.

Clearly, the type of disinfectant played a more important role than BAC-filtration in controlling the consumption of DO in this study. Of course, chlorine is known to be a better disinfectant than chloramine., which can account for the lower DO consumption in the chlorine Pipe reactors when residuals were still present. Other possible factors for higher DO after the disinfectant residual disappeared include production of DO up to 0.95 mg DO from 4 mg free Cl_2 auto-decomposition[50,51,52] and cytotoxicity from DBPs produced by free chlorine reactions with organic matter inhibiting biological activity.[53] The lower levels of turbidity and sediment in the chlorine rigs may also translate to less microbial activity in the sediment, while nitrification in the presence of chloramine also consumes oxygen (discussed further in section 3.4).

3.3.3 Rapid disinfectant and DO loss in Tube Rigs

Despite the dramatic difference in reactor characteristics and sediment accumulation, there was also very rapid loss of disinfectant and DO in the Tube rigs. Specifically, both chlorine and chloramine were almost depleted by the end of the 15.24-meter (50-ft) tubes after only a 32-min hydraulic residence time. In contrast to the Pipe RWDS rigs, where chloramine was depleted faster than chlorine, chloramine was more persistent than chlorine in the Tube rigs. In samples collected at the end of the study, about 50% of the chlorine residual was lost at the 3.05-meter (10-ft) tubing interval, compared to just 3% of chloramine.

At the end of the Tube rigs, chlorine was non-detectable, whereas chloramine was still measureable at 0.24 mg/L (Figure 9a).

The DO level after just 32-min residence time in the Tube rigs was roughly equivalent to that observed at 1,440-min (1-day water age) in the Pipe rigs. This difference factor of roughly 45 (1440/32) likely reflects the greater SAV ratio for the Tube versus Pipe RWDS (32 × greater), enhancing the influence of bio-foulant growth on bulk water chemistry. The DO trends in the Tube rigs were similar to those observed for the Pipe rigs with respect to the type of disinfectant. The DO drop decreased on the order of 6.22 – 8.92 mg/L, > 3.73 – 4.17 mg/L, and > 0.95 – 1.21 mg/L for the chloramine, no disinfectant, and free chlorine conditions, respectively (Figure 9b). Disinfectant decay and DO trends were consistent in the duplicate Tube rigs (Fig S2 and S3).

3.4 Rapid chloramine decay and nitrification

In the present study, BART testing of the filtered and unfiltered conditions revealed very high levels of nitrifiers in the bulk water and sediment of the first pipe segment in the chloramine condition (Table S1). The semi-quantitative BART results were all over range in all four (filtered/unfiltered sediment and water) chloramine samples (>10^5 cfu/ml), which helps explain the observed rapid chloramine loss in this segment. This is consistent with an analogous study of simulated DWDSs using similar Pipe rigs, where higher than expected chloramine loss rates were attributed to nitrification.[14] Another study also reported "super nitrification" and high levels of ammonia-oxidizing and nitrite-oxidizing bacteria near the entry point of a simulated RWDS.[54] Bal Krishna et al.[55] further showed that the presence of soluble microbial products, which were likely to be abundant in the water and sediment in the chloraminated condition in this study, can further accelerate the rate of nitrification. There

were much lower levels of nitrifiers (10^3 cfu/mL) in the unfiltered no disinfectant residual Pipes. Even though the influent water underwent breakpoint chlorination to remove ammonia, ammonia can be generated during microbial degradation of organic matter, e.g., as observed in lake sediment.[56,57] No other conditions had detectable nitrifiers (Figure S4).

HPCs measured in sediment and bulk water samples were very high in the first pipe segments of the chloramine and no residual Pipes, ranging from 5×10^5 to 1×10^6 cfu/mL. This is consistent with other indications of high biological activity in these conditions. Remarkably, no HPCs were detected in either chlorine bulk water or sediment, even on plates inoculated with undiluted samples. This confirmed that the superchlorination pretreatment without BAC-filtration, or a BAC-filtration with 4 mg/L chlorine residual, dramatically reduced biological regrowth. This was consistent with the low sediment VS/TS ratio (Figure S5).

3.5 Flask test comparing disinfectant demand for Pipe RWDS sediment versus biofilm

The disinfectant decay trends observed within the Pipe and Tube RWDSs represent a composite of reactions occurring in the bulk water, biofilm, and sediment under defined flow regimes. A batch flask test was designed to elucidate the relative contributions of each component to the total observed disinfectant demand. Chlorine decay in all test conditions generally fit first-order decay kinetics, with moderate to high R^2 values ranging from 0.79 to 0.98 (Table S2). In the filtered conditions with chlorine, the reactions in bulk water alone without sediment or biofilm resulted in about 50% residual loss in 24 hours. However, addition of swabbed biofilm achieved a 50% chlorine loss 4 – 9 times faster (i.e., 2.5 – 6 hours) indicating high free chlorine demand from biofilms (Figure 10). Addition of sediment from this

system did not appreciably increase the rate of chlorine loss beyond that observed in the bulk water alone. Sediment in the chlorine pipes versus chloramine pipes contained 16.7-44.1% less VS (3.6-10 g/L vs 21.5-22.7 g/L) and a lower ratio of VS/TS (48-51% vs 79-83%) (Table 1), indicating less biologically active sediment on an absolute and relative basis. In contrast to the rapid chlorine loss, chloramine was extremely stable in the BAC-filtered bulk water condition, with more than 6 days required to achieve 50% residual loss. However, in the presence of biofilm or sediment, chloramine loss was 6.8 – 11 times faster, achieving 50% loss in 5.5 hours with sediment addition and in 20 hours with biofilm addition. For the corresponding unfiltered chloramine conditions, 10 – 10.5 hours was required to achieve a 50% residual loss in the bulk water or biofilm condition, but with sediment chloramine loss was about twice as fast, achieving 50% loss in only 5.5 hours. This is consistent with several indications of high biological activity in the sediments from the chloramine rigs and the observation that it was rich in nitrifiers that can cause a high chloramine demand. The above results also help explain the difference in relative stability of chloramine versus chlorine in the Tube versus Pipe rigs. The Tube rig design prevented accumulation of the sediment that was the major source of chloramine demand in the Pipe rig, but the high SAV ratio enhanced the effect of biofilm that was a major source of chlorine demand. The net result is that in the Tube rigs, chloramine was more stable than chlorine (Figure 9a), whereas in the Pipe rigs the opposite trend was observed (Figure 7a).

The only other lab-scale simulated RWDS study comparing chlorine versus chloramine disinfectant residuals used a Tube rig design with the same diameter tubing (0.318-cm) and reported very rapid free chlorine loss of 0.04 – 3.6 mg/L (average 1.23 mg/L) with just a 4-min

hydraulic residence time.[6] In that study, the chloramine loss rates were lower, at 0.09 – 1.23 mg/L (average 0.54 mg/L), in agreement with the results presented herein.[5]

3.6 Formation of DBPs in the Pipe Rigs

DBP formation in chlorinated and chloraminated distribution systems is a topic of ongoing research in DWDSs.[58, 59] Although there are no DBP regulations for reclaimed water,[60] regulated drinking water DBPs were examined in the six Pipe RWDS reservoirs to assess the maximum DBP-generating potential in each condition. After superchlorination in the case of unfiltered chlorine and breakpoint chlorination for all other conditions, and then 30 minutes reaction time with the targeted residual type and dose, all DBPs exceeded the 80 ppb total trihalomethane maximum contaminant level drinking water standard (Figure 11).[61] The highest levels of DBPs were measured in the two Pipes with chlorine residuals, with chloroform dominating. Levels of regulated DBPs in the chloramine Pipes and in the no disinfectant residual Pipes were comparable. Chloraminated DBPs were not measured in this study.

4. Conclusion

While it is not possible to precisely replicate a real-world RWDSs in the lab, the designs implemented in this study can provide insight into expectations representing extremes encountered in real-world systems. Findings here can aid in identifying management strategies that minimize potential for biofouling and sediment accumulation in RWDSs, and improve understanding of how distribution of reclaimed water could alter its suitability at the point of use.

Pipe versus Tube RWDS rigs were employed to represent extremes in hydraulic regimes and potential to accumulate sediment. Pre-treatments, disinfectant types, and hydraulic design

controlled the amount and nature of sediments that accumulated. Sediment deposits in systems with chloramine or no disinfectant residual were highly biologically reactive compared to those in systems with chlorine, as indicated by VS/TS ratio, HPCs, disinfectant and DO decay.

The relative importance of sediment versus biofilm in Pipe versus Tube RWDS rigs and dominance of biofilm surface area determined the relative stability of chlorine versus chloramine residual. Specifically, the large diameter and very low flow velocity led to accumulation of sediment at the bottom of the Pipe rigs, resulting in rapid loss of disinfectant residuals and dissolved oxygen with depth near the entry point. The biologically active sediment was the main source of chloramine disinfectant demand and was a hotspot for nitrification. Severe bio-fouling was observed in the Tube rigs with chloramine and no residuals. The high biofoulant SAV ratio in the chloramine tubes dominated the decay and resulted in rapid depletion of the disinfectant. Much less sediment and biofouling were observed in the chlorinated Pipe and Tube rigs, where biofilm was the main source of disinfectant demand.

This work demonstrated that sediment and/or bio-foulant accumulations are both important features of RWDSs, which in turn can control the interplay between chemistry and microbiology. The discovery of significant sediment accumulation in RWDSs has important implications for monitoring and managing nitrification when chloramine is used as secondary disinfectant. Sediment accumulation may likely foster growth of opportunistic pathogens in RWDSs, as has been reported to be the case for potable water systems, [7,8] which are much less susceptible to accumulation of sediment. The differing origins of disinfectant demand for chloramine versus chlorine also reveals a new dimension to the conceptualization of water quality deterioration during transport though RWDSs.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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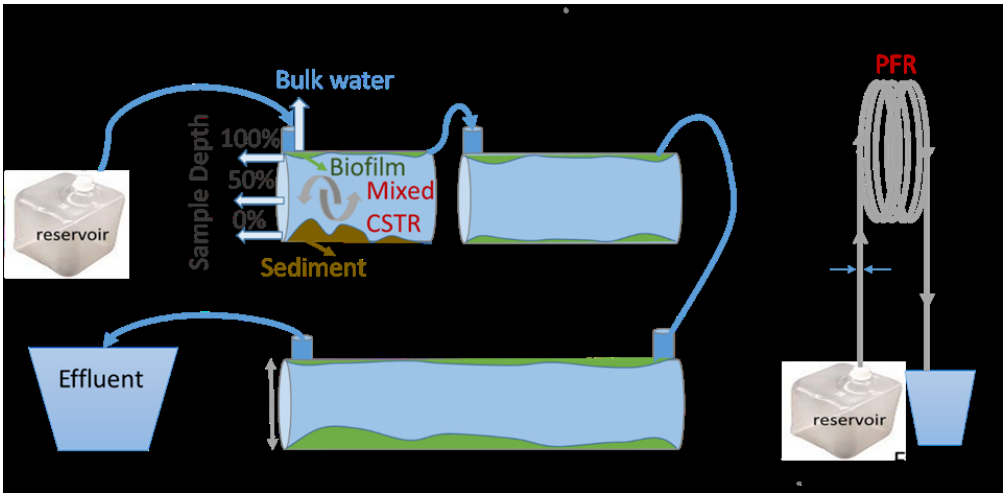

Figure 1 (a) Simulated 10.16-cm (4-in) Pipe RWDS rigs with sampling ports at the point of entry (P0), and at calculated water ages of 1-Day(P1), 2.5-Day (P2), and 5-Day (P3). Samples were also taken at the bottom (0%), mid-point (50%), and the top (100%) of the vertical depth of the pipe. (b) 3.175-mm (1/8-in) Tube RWDS rigs with total length of 15.24-m (50-ft) and sampled every 3.05-m (10-ft). Duplicate Tube rigs were set up in parallel for each disinfectant condition to compare reproducibility.

162x79mm (150 x 150 DPI)

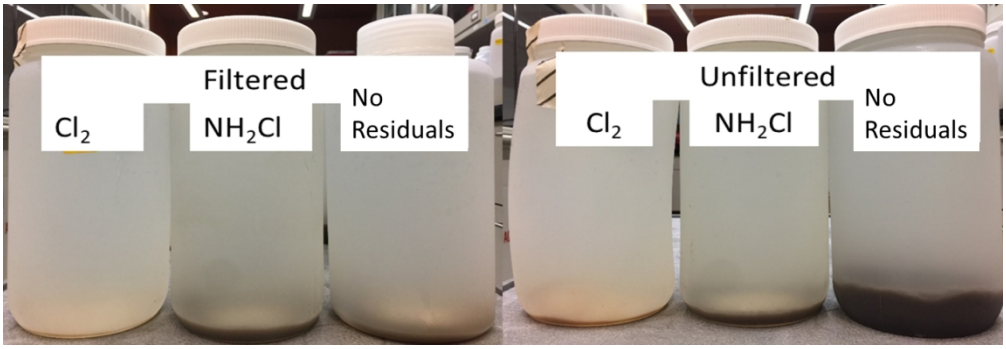

Figure 2 Settled sediment removed from the bottom of the first pipe segment of the six Pipe RWDS conditions (2-L containers). Sediment was collected after three years of operation, except for the unfiltered chloramine condition, which was collected after two years of operation due to a pipe failure and replacement mid-experiment.

236x80mm (150 x 150 DPI)

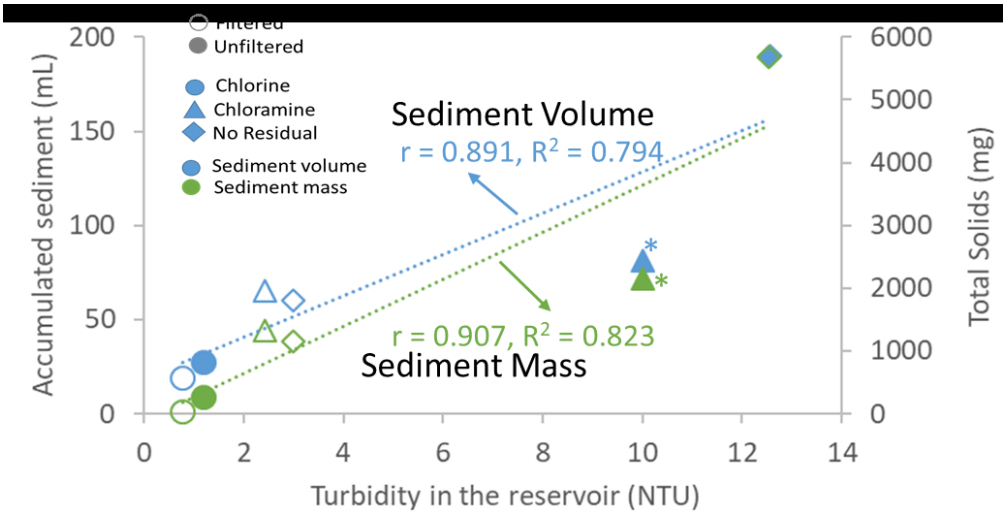

Figure 3 Correlation between the turbidity of the influent reservoir and accumulated sediment in the first pipe segment of the Pipe RWDS after 39 months of operation, as measured by sediment volume (blue symbols, primary y-axis) and total solids mass (green symbols, secondary y-axis). The solid symbols in the legend indicate unfiltered influent and the empty symbols indicate BAC filtered influent. * Due to a pipe replacement, 39 months accumulation was linearly extrapolated based on measurements after 24 months of operation.

174x89mm (150 x 150 DPI)

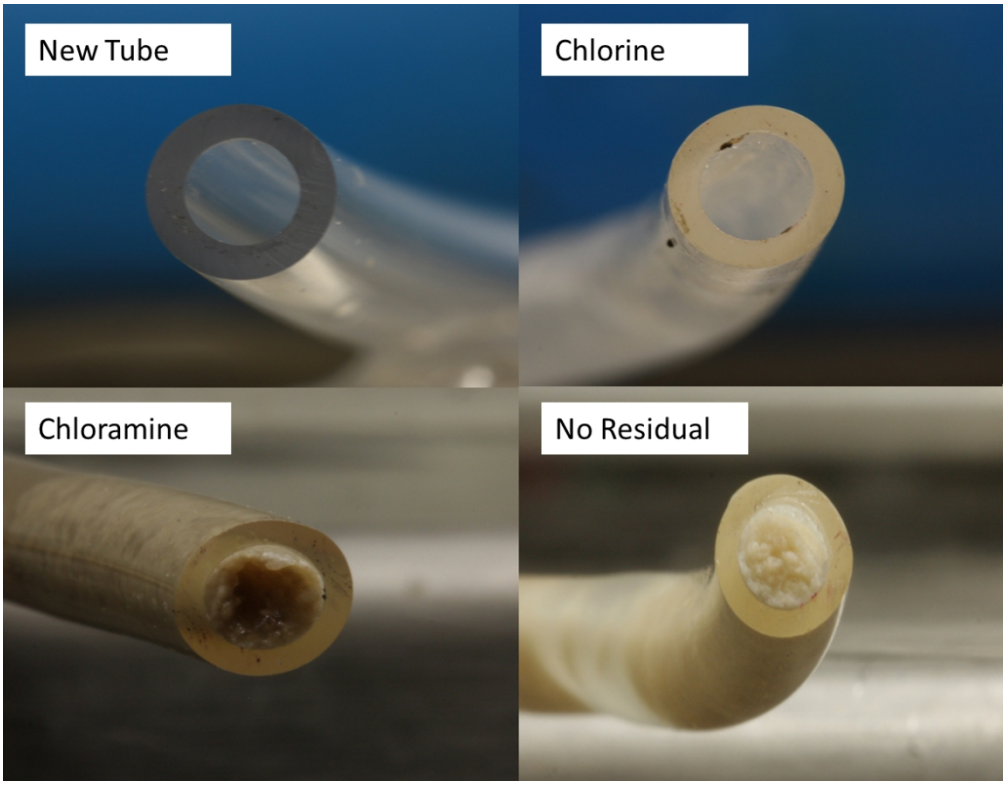

Figure 4 Cross-section photographs of the Tube rigs revealed that the extent and morphology of sediments/biofilms were distinct across the three disinfection conditions. Photos were taken at cross sections 40-ft from influent. Only unfiltered feed water was used to prepare the three disinfectant conditions in the Tube Rigs.

198x153mm (150 x 150 DPI)

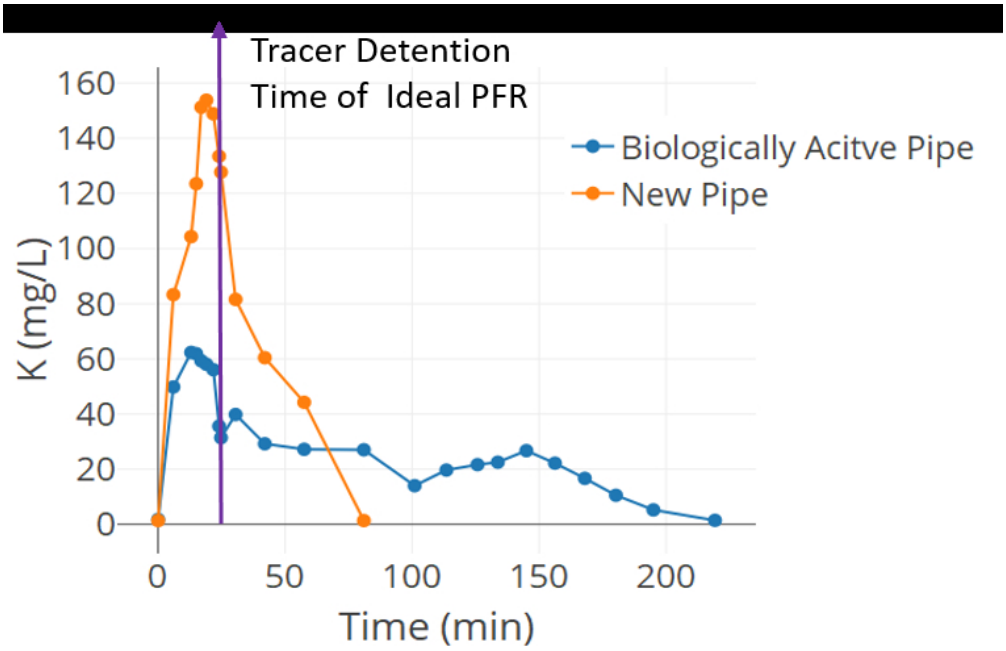

Figure 5 Tracer study in the first pipe segment of new versus biologically-active (i.e., operated ~3 years) Pipe Rigs. Potassium (K) was injected at 50,000 mg/L potassium chloride. The ideal plug flow reactor (PFR) wash out curve is shown for comparison.

128x83mm (150 x 150 DPI)

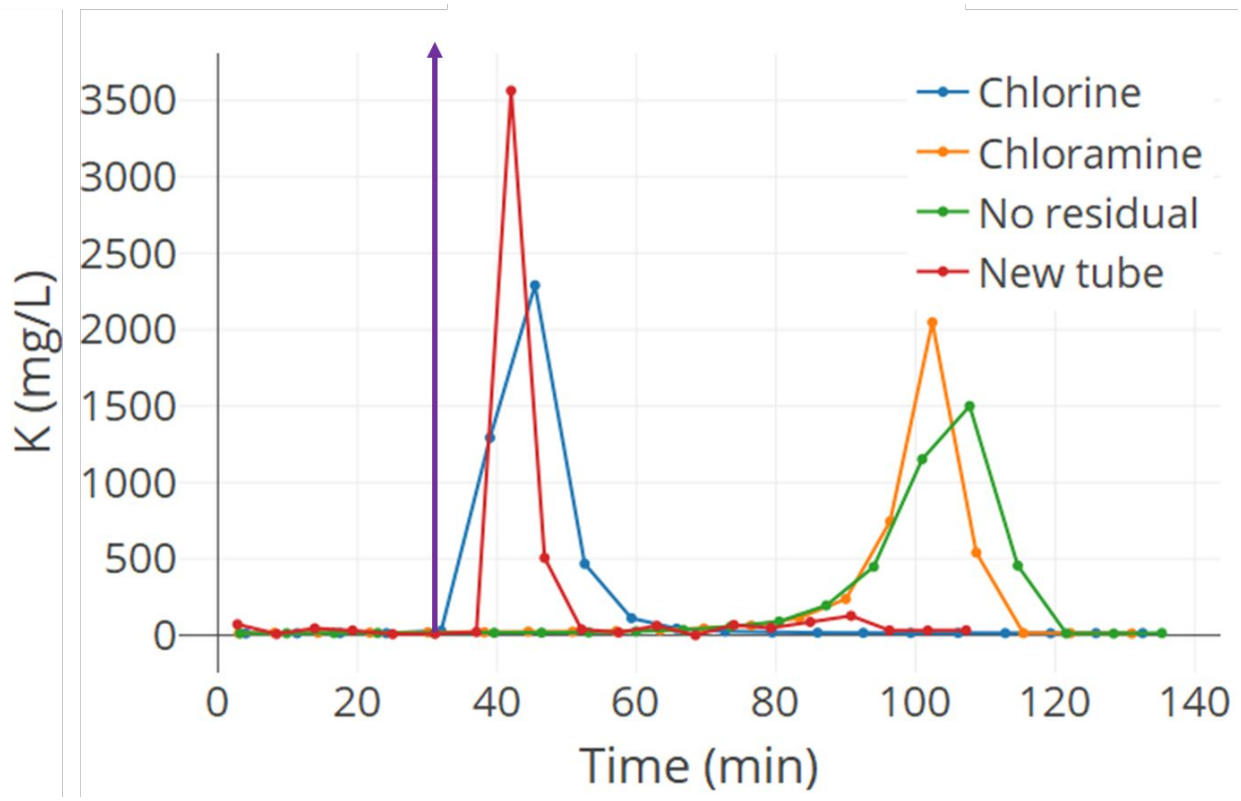

Figure 6 Potassium tracer studies in the Tube rigs demonstrated PFR flow but with a delayed retention time for conditions with heavy biofilms. Potassium (K) was injected at 30,000 mg/L KCl.

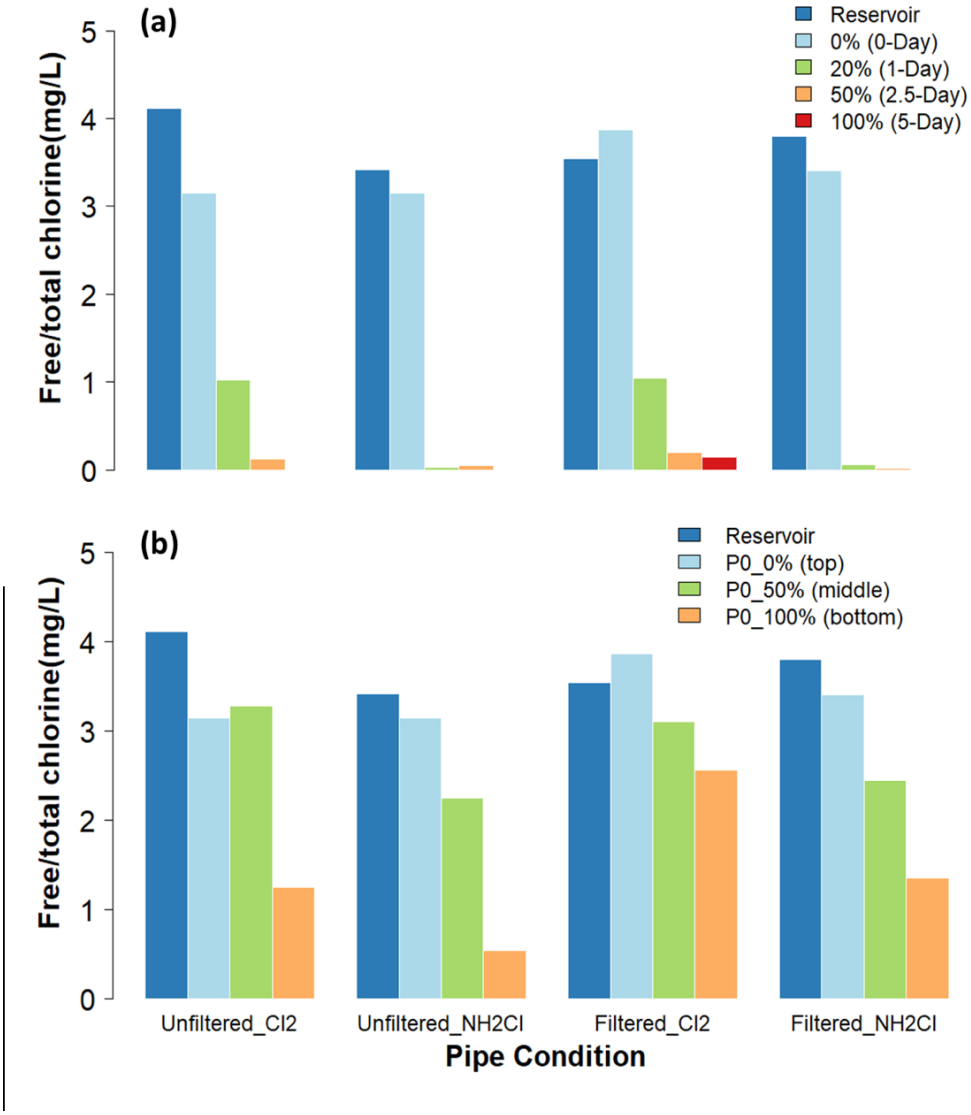

Figure 7 Depletion of disinfectant residuals (a) along the horizontal pipe flow direction and (b) with increasing depth at the point of entry (P0) of the Pipe RWDS. The a and b sampling events were conducted 1 week apart at an operational temperature of 22 ^{0}C. The legend in (a) indicates calculated hydraulic residence times (i.e., assuming plug-flow for an empty Pipe) at the sampling ports and in (b) percent distance from the top of the Pipe. Measurement in (a) were sampled in reverse order from the last sampling point (P3) to the first sampling point and measurements in (b) from the top to bottom water level to minimize disturbance of subsequent sample. Filtered = BAC filtered influent; Unfiltered = unfiltered influent; Cl2 = chlorine condition; NH2Cl = chloramine condition.

130x150mm (220 x 220 DPI)

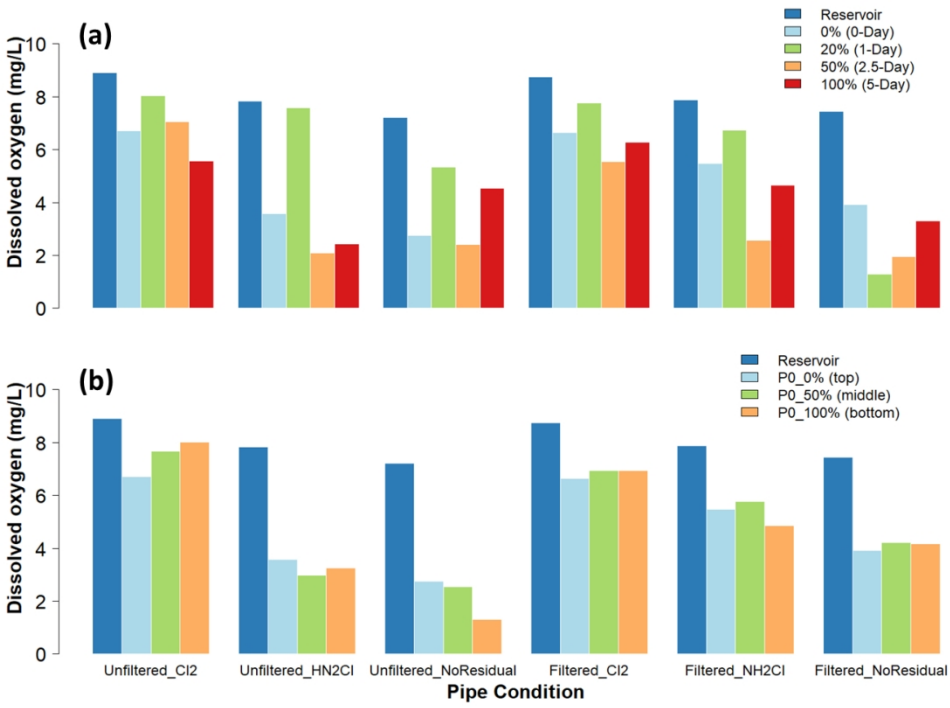

Figure 8 Dissolved oxygen level (a) along the horizontal pipe flow direction and (b) with increasing depth at the point of entry (P0) of the Pipe RWDS. The legend in (a) indicates calculated hydraulic residence times (i.e., assuming plug-flow for an empty Pipe) at the sampling ports and in (b) percent distance from the top of the Pipe. Measurements in (a) were sampled in reverse order from the last sampling point (P3) to the first sampling point and measurements in (b) from the top to bottom water level to minimize disturbance of subsequent sample.

232x174mm (150 x 150 DPI)

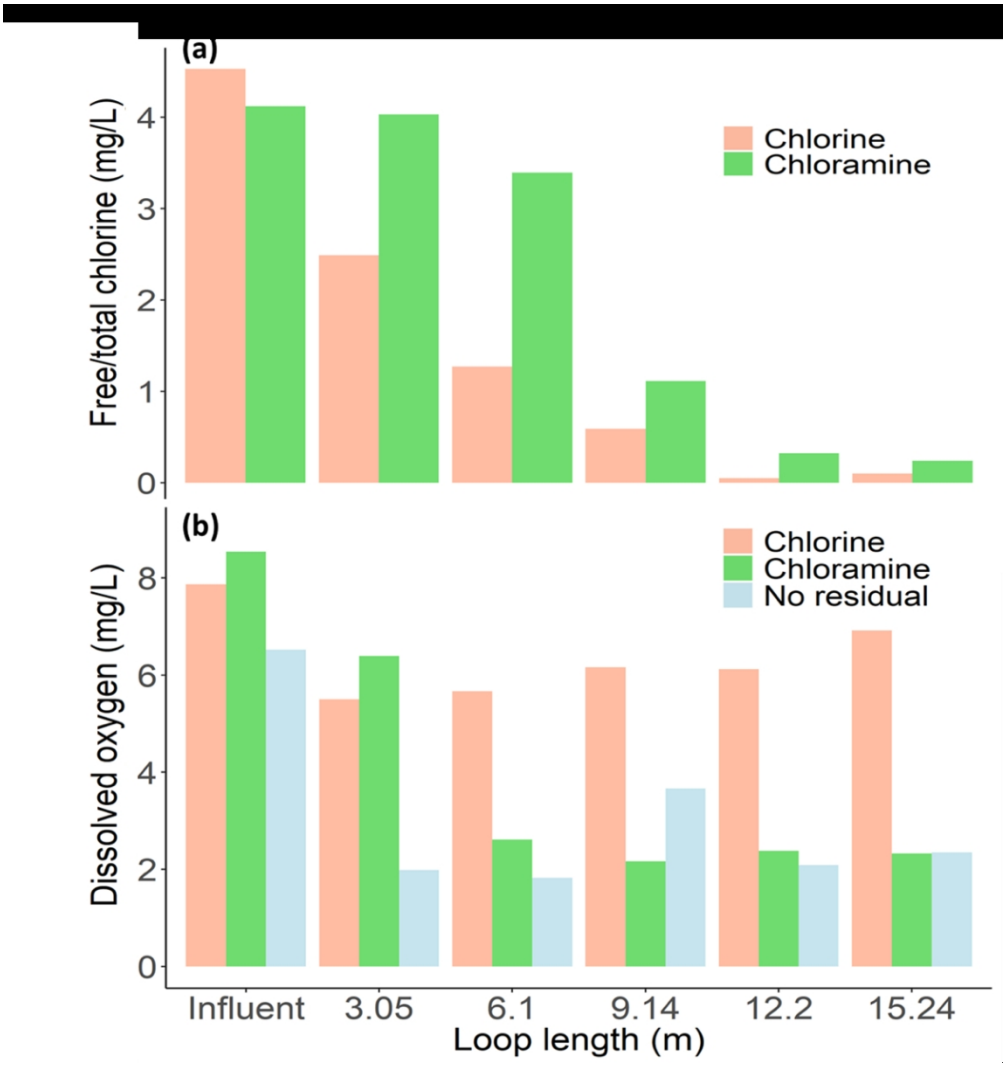

Figure 9 (a) Chlorine and chloramine measured at the end of the 15.24-m (50-ft) Tube rigs (31-min calculated residence time) and corresponding (b) dissolved oxygen measurements at 3.05-m (10-ft) intervals of the Tube rigs, measured by cutting and sacrificing the tubing at the end of the experiment.

138x146mm (220 x 220 DPI)

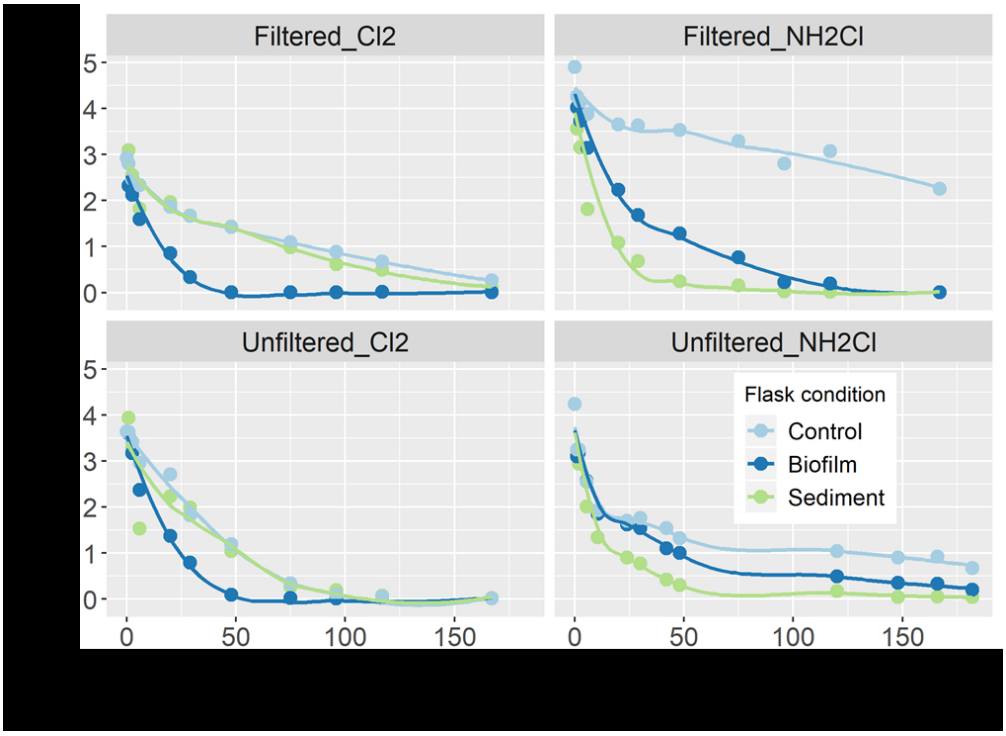

Figure 10 Disinfectant decay curves observed in batch-scale flask study. 10 mL sediment or a biofilm swab of 10 cm2 from the indicated Pipe RWDS conditions was added to 250 mL flasks containing a targeted initial chlorine or chloramine residual of 4 mg/L added to biofilm/sediments collected from the corresponding rig from which it was collected. Cl2 = chlorine; NH2Cl = chloramine; filtered = BAC-filtered influent; unfiltered = no BAC pre-treatment.

171x124mm (150 x 150 DPI)

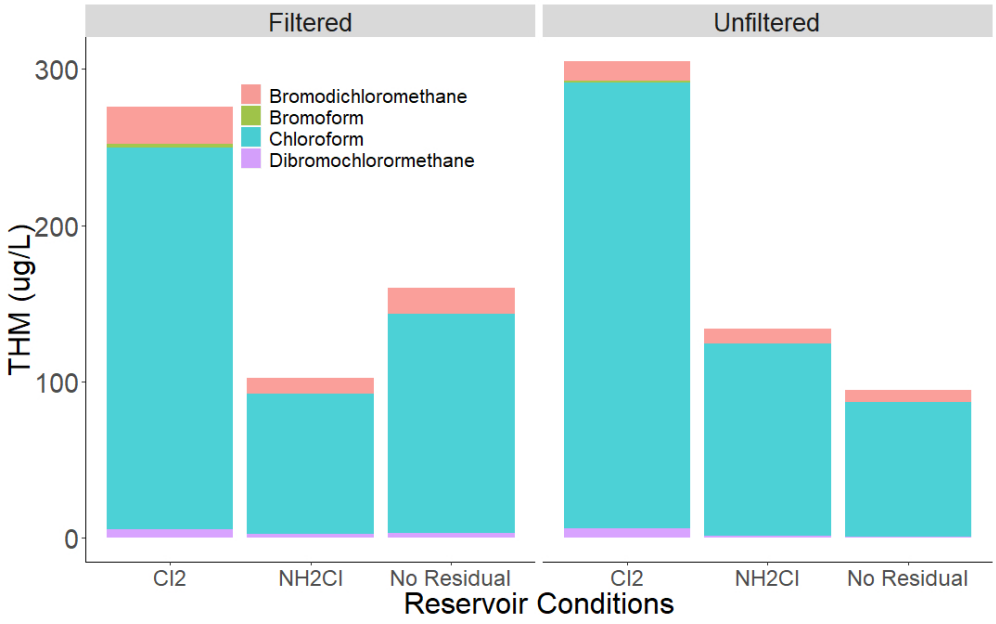

Figure 11 Illustrative trihalomethane (THM) levels in the six reservoirs, 30 minutes after adding chlorine or chloramine. As there were no detectable THMs in the treatment plant effluent, THMs in the condition with no residual reflect prior breakpoint chlorination.

142x89mm (192 x 192 DPI)

Table 1 Summary of key water quality parameters of the two feed water conditions, before and after BAC-filtration, and the six influent conditions representing different BAC-filtration and disinfection strategies.

	Reservoir Conditions					
	Unfiltered			**BAC-Filtered**		
TOC (mg/L)[a]	6-8			<2		
Soluble AOC[b] (ppb)	48.6-160			11.9-29.3		
Total AOC[b] (ppb)	399-610			292-660		
Total cell counts[c] (TCC) (cells/µL)	6624 ± 2190			2672 ± 1136		
Chlorine demand (mg/L)	2.85 ± 1.87			1.16 ± 0.0006		
	Six Influent Conditions					
	Chlorine	Chloramine	No Residual	Chlorine	Chloramine	No Residual
Targeted residual (mg Cl_2/L)	4 as chlorine	4 as chloramine	No residual	4 as chlorine	4 as chloramine	No residual
Mean residual[d] (mg Cl_2/L)	3.81	3.92	0. 01	3.69	3.87	0.02
Total NH3 (mg/L)	No	0.98	No	No	0.97	No
Breakpoint chlorination	Yes	Yes	Yes	Yes	Yes	Yes
Superchlorination	Yes	No	No	No	No	No
BAC-filtration	No	No	No	Yes	Yes	Yes
Turbidity[e] (NTU)	1.20	10.0	12.6	0.78	2.43	3.00
Sediment volume (mL)	27.0	50.0*	190.0	19.00	65.0	60.0
Sediment/Pipe volume ratio (%)	0.45	0.83	3.2	0.32	1.1	1.0
Volatile Solids (VS) in Sediment (g/L)	10.0	22.7	24.9	3.63	21.5	19.5

Total Solids (TS) in Sediment (g/L)	21.0	27.4	32.5	7.13	26.3	25.6
Mass of VS (mg)	121	793	4366	14.5	1074	879
Mass of TS (mg)	252	957	5679	28.5	1313	1150
VS/TS (%)	48.1	82.8	76.9	50.9	79.1	76.4

***** Sediment collected in the unfiltered chloramine Pipe only corresponded to Sep 2016 – Sep 2018 due to pipe replacement.
a The range of TOC values were collected over the 3-year operation.
b Soluble and total AOC range were from 2 independent measurements at the beginning of the final sampling event.
c Total cell counts measurements were averaged over 6 sampling events throughout the 3-year operation.
d The mean disinfectant residuals in the influent reservoirs were averaged over 245 measurements collected at each water change over the 3-year operation.
e Turbidity was measured as a one-time event at the final sampling event.

Sediment and Biofilm Affect Disinfectant Decay Rates During Long-term Operation of Simulated Reclaimed Water Distribution Systems

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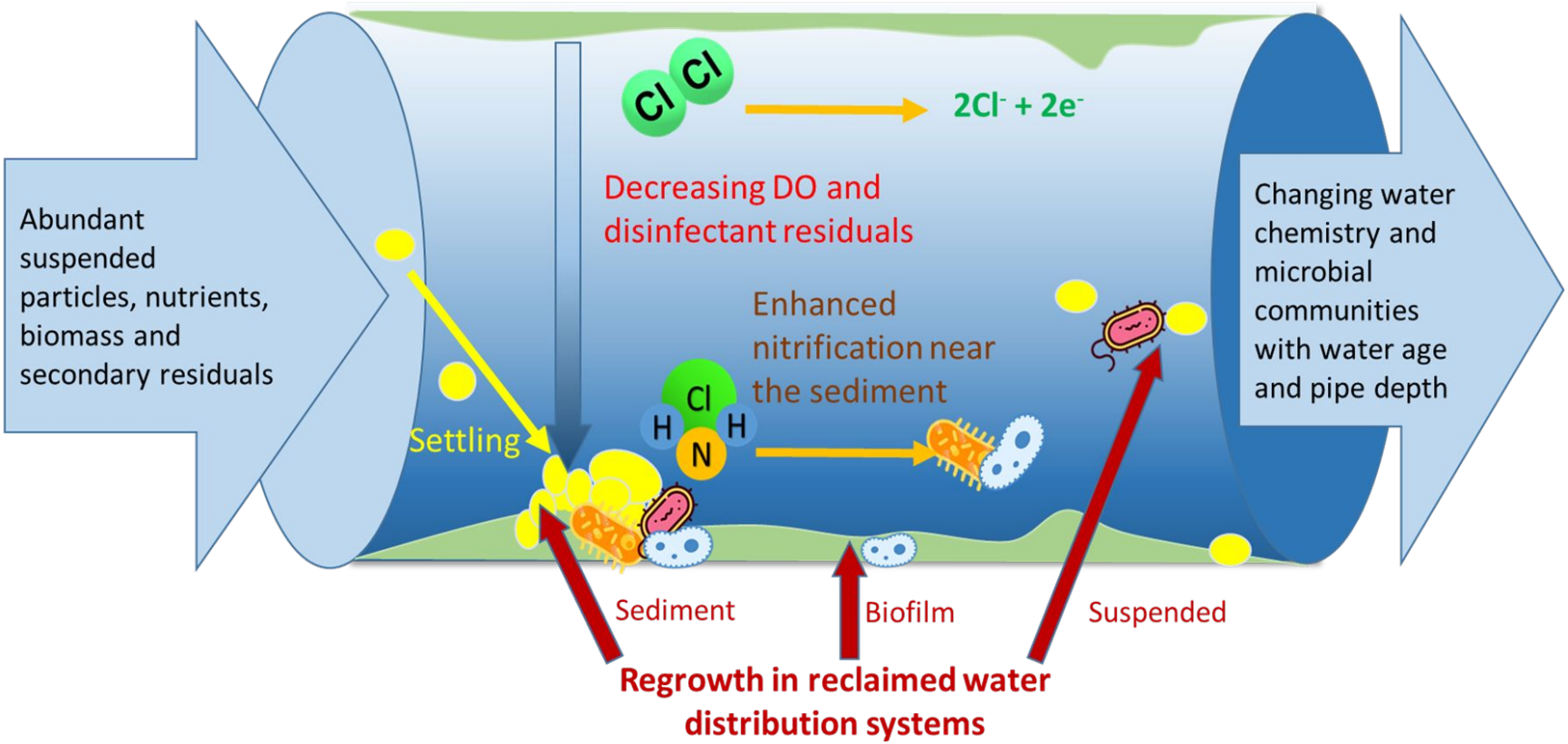
Table of Content



Unique water chemistry and operational conditions of reclaimed water distribution systems facilitated accumulation of sediment which resulted in depletion of disinfectants and microbial regrowth.