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Biofilm sloughing and CBP formation in a 12-km transport system carrying chlorinated saline secondary effluent

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ABSTRACT

A 12-km conveyance system has been placed in service for 10 years to transport treated municipal saline effluent which was not chlorinated. Now the effluent is being considered for chlorine disinfection to reduce *Escherichia coli* count to less than 1,000 cfu/100 mL. Since the system was not designed to carry the chlorinated flow, a simulator study has been conducted with a 68-m pipe loop (2.5″ ID) to assess the biofilm growth and sloughing when the effluent is subjected to chlorination. In addition, the potential formations of THM and HAA_5 in the transport system are also assessed. Experimental results indicate that the level of biofilm growth in the pipe wall is mainly dictated by the organic strength of the effluent flow. For conveying the saline secondary effluent with a BOD of about 10 mg/L, the biofilm growth is approximately 160 µm. With chlorination of up to 5.0 mg/L, the biofilm growth and sloughing are not affected. As for CBP productions, 5.0 mg/L chlorination of the activated sludge effluent (with an operating sludge age of 5–12 days) does not result in any excessive CBP formations. The maximum THM and HAA_5 levels ever found in the chlorinated effluent are only 40 and 10 µg/L, respectively. This is because the added chlorine reacts instantaneously with effluent ammonia to form much less reactive combined residuals.

Keywords: Chlorination of saline secondary effluent; CBP formation; Biofilm growth and sloughing in force main

1. Introduction

Discharge of non-disinfected municipal effluent has often caused much public concern particularly when the receiving water is used for certain water contact activities such as boating and swimming. More and more stringent standards have been promulgated in recent years in order to ensure better public health [1]. The conventional biological processes such as activated sludge and trickling filters are known to remove up to 90–99% of some microorganism; however, this is not sufficient to guarantee the safety of public health since the coliform level in the treated effluent is still high and the receiving water may not be suitable for bathing and other activities [2]. In many cases, tertiary treatment may be needed to further remove suspended solid (SS), turbidity, color, offensive odor, organic matter, pathogens and viruses [3]. Thus, effluent disinfection is now often included in the

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municipal sewage treatment chain. The required level of disinfection is normally dictated by the promulgated standards for a specific designated area [1].

A wastewater treatment plant normally has an effluent transport system to deliver the treated sewage to its receiving water. In some cases, such a system may also be used as a "contact chamber" upon chlorination. The biofilm ecosystem in an effluent pipeline is quite complex and poorly understood because it is sensitive to the change of physicochemical conditions in the flow [4]. The term "biofilm" refers to a thin layer of microbial community adhered to the conveyance pipe wall [5]. There are several factors affecting the biofilm development, including microbial nutrients, concentration of biodegradable organic matter, pipe materials (or roughness), temperature, disinfectant residual, bacteria from the carried flow, shear stress at the biofilm–liquid interface and the hydraulic regime, etc. [6]. Among these, the key controlling factors may be linked to the levels of biodegradable organic matter and the disinfectant residuals [7]. The biofilm and its trapped organic matter have a high reducing potential, which can lead to a high chlorine demand. This of course will result in some chlorine decay and lower the disinfection performance in the conveyance system when it is used as a contact chamber. At the same time, through chlorine oxidation, a portion of biofilm and its trapped organic matter may become detached and returned to the carried water [8].

Different sludge ages in the activated sludge operation may result in different effluent qualities. This may in turn impose some variations in chlorine demand and CBP productions. At low temperature, a long sludge age is commonly adopted to maintain adequate nitrification. In general, the effluent-derived organic matter (EfOM) in activated sludge operation is not highly aromatic, and it is often regarded as bio-stabilized and less reactive with chlorine than the assimilated organic matters derived from natural sources [9]. However, EfOM is rich in dissolved organic nitrogen, which may serve as precursors for the CBP formations, including trihalomethanes, haloacetic acids, haloacetonitriles, haloacetaldehydes, and nitrosamines, etc. [10].

In one wastewater treatment system in Hong Kong, its existing effluent transport system consists of 5-km ductile iron force main and 7.2-km concrete tunnel. This system has been in service for over 10 years to transport non-chlorinated secondary effluent. At present, chlorination for the secondary effluent is being considered to reduce its *Escherichia coli* to below 1,000 cfu/100 mL in order to improve the bacteriological water quality in its receiving harbor water. However, the growth and sloughing of biofilm in the conveyance system carrying the chlorinated flow and the resultant impacts on the effluent water quality, such as SS increases, CBP formations and iron release from the metal corrosion of force main, etc. are largely unknown. Thus, before chlorination implementation, there exists some practical concerns, including: (1) will a heavy slough-off of biofilm occur such that the SS level in the carried flow is greatly increased; (2) will a high production of CBP take place in the transport system; (3) will the sludge age of activated sludge operation have a bearing on the CBP formation; and (4) will the chlorinated flow accelerate the corrosion of existing ductile iron force main. To ease the above concerns, a simulated study has been conducted to find the answers to the above questions. This paper will present data to address the first three concerns.

2. Materials and methods

2.1. Set-up of a simulated pipe-loop system

In order to assess the biofilm slough-off and its subsequent impact on the effluent quality upon chlorination, a specially designed 68-m pipe loop is constructed with commercial 2.5-inch PVC pipe. The loop system includes 12 detachable sections (each 20-cm long), each containing two stainless steel strips (each 2.0-cm wide) which are clasped firmly to and flushed with the inner pipe wall. These strips are removable so that the changes in biofilm growth (thickness and weight) before and after chlorination can be measured. The total volume of the pipe loop is 233 L. During operation, about 500 L of the actual saline effluent, which was collected from a municipal sewage treatment plant, is placed in a 1,000-L feeding (polyethylene) reservoir. A centrifugal pump is used to deliver the flow to the simulated pipe loop at a controlled flow rate while chlorine is injected at the pump's discharge end.

2.2. Cultivation of biofilm growth in pipe loop

In order to speed up biofilm development in the pipe loop, primary effluent from the actual treatment plant was used to circulate through the simulator loop. Besides, about 150 mg/L sugar COD was also added to the primary effluent in the first month of operation. Similar to the finding by Ref. [11], a steady-state biofilm growth was observed after four weeks of operation. The biofilm thickness reached a level of about 1,110 μm. The corresponding biofilm dry density averaged about 29.4 mg/cm^3. The VSS/TSS ratio in biofilm was around 0.60–0.65, indicating that the biofilm had a sludge age of at least 20 days. Upon reaching the full biofilm development, the feeding was switched to the actual secondary effluent without any sugar supplement. Subsequent to this, the biofilm thickness was progressively declining, and it eventually reached a new steady state level of about 160 μm.

2.3. Biofilm measurement

There are different methods for measuring the biofilm thickness [12, 13]. In this study, the methodology of using a modified micrometer and a microscope, as described by Ref. [14] was employed for the biofilm measurement. It was quite simple and reasonably accurate. After each removable S/S strip was taken out from the pipe section, the excess water was allowed to drain for 5 min. Then the strip was mounted on the stage of a microscope. The microscope was first focused at the biofilm surface. After the sharp tip of the micrometer just touched the biofilm surface, the first reading on the micrometer was taken. Then the tip was forced through the biofilm to touch the metal surface. At this time, the second reading was taken. The difference of these two readings gave the biofilm thickness. Since the thickness varied from one place to another, normally 10–15 thickness measurements were taken at random from each strip. The average value of these measurements was used for data calculation.

2.4. Batch test for CBP production

To assess the CBP production in the chlorinated effluent, the non-chlorinated saline effluent samples were placed in several 2-L glass beakers. The effluent was first mixed by a magnetic stirrer. During mixing a predetermined level of chlorine (2.0 and 5.0 mg/L as Cl_2) was dispensed to the sample. Each test was allowed to have a contact time of 60 min which equals to the actual flow travel time in the 12-km conveyance system. The solution pH was unadjusted. After each test, the chlorinated effluent was taken for two types of analyses: (1) chlorine residuals (including free chlorine and monochloramine); and (2) CBP formations including THMs ($CHCl_3 + CHCl_2Br + CHClBr_2 + CHBr_3$) and HAA_5 ($ClCH_2COOH + Cl_2CHCOOH + Cl_3CCOOH + BrCH_2COOH + Br_2CHCOOH$). For the CBP determinations, each collected sample was immediately de-chlorinated with sodium bisulfite to neutralize any remaining chlorine residuals in the sample.

2.5. Chemical analyses

Suspended solid measurement was conducted following the Ref. [15]. Chlorine residuals were measured by DPD/FAS titration, and THMs and HAA_5 were measured by a gas chromatograph with electron capture detector (GC/ECD) after extraction in accordance to the USEPA Method 551.1 [16]. All analyses were performed immediately after each sample collection. In assessing the CBP formation, the collected sample was first subjected to extraction. The extracts, if not analyzed immediately, were kept in a 4°C cool room. In all cases, the CBP analyses were completed within 3 days from the sample extraction. For QA/QC, at least one duplicate was made for every 10 samples taken. This was to ensure that the test results were always consistent.

3. Results and discussion

3.1. Effect of chlorination on biofilm growth and sloughing

During the period of feeding the secondary effluent, the biofilm buildup in the pipe loop was frequently monitored. Before any chlorine application, the average biofilm thickness was 157 μm, and its dry volumetric density was 20.6 mg/cm^3. These data were observed on the 80th day of the loop operation.

Upon the first chlorine application on the 82nd day, the progressive decline of chlorine residuals with time after 2.0 mg/L chlorine was dosed to the pipe loop is shown in Fig. 1. It is worth pointing out that the 2.0 mg/L chlorine dosing was able to reduce *E. coli* to below the 1,000 cfu/100 mL limit. After chlorine dosing, the first sample was taken at a contact time of 4 min. Its total residual was only 0.30 mg/L, one half of which was free and the other half was the combined residual. The combined residual continued to decline and reached 0.05 mg/L at the end of 60th min. In order to allow the biofilm to be exposed to the combined residual for a longer time, another 0.20 mg/L of mono-chloramine was added at the 62nd min. After this, the combined residual was found to be only 0.15 mg/L at the 64th min. The residual continued to decline to 0.05 mg/L at the end of the 120th min (Fig. 1).

In this study, it was found that even under continued exposure to chlorine, the biofilm did not show any

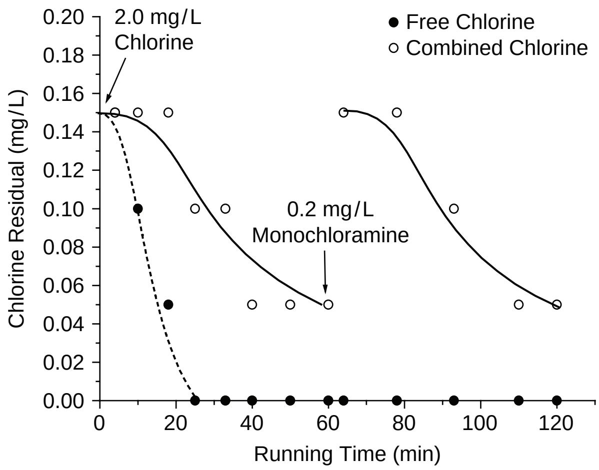

Fig. 1. Decline of chlorine residuals with time after 2.0 mg/L chlorine dosing on the 82nd day.

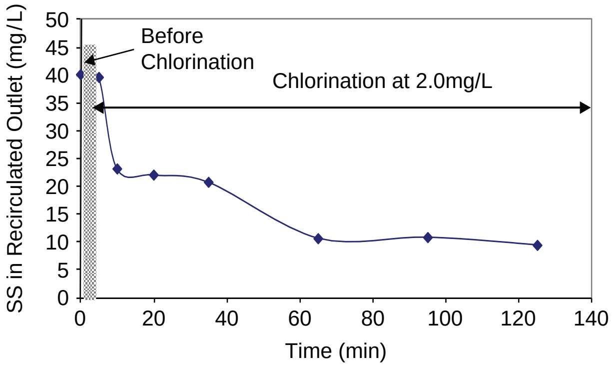

Fig. 2. Observed SS level at the pipe loop exit after chlorination at 2.0 mg/L on the 82nd day.

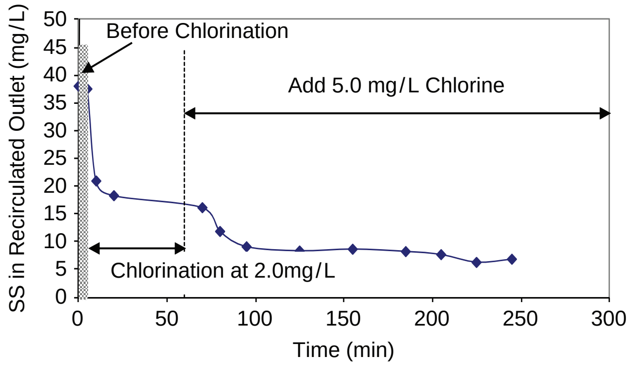

Fig. 4. Observed SS level at the pipe loop exit after chlorination at 2.0 and 5.0 mg/L in sequence on the 83rd day of operation.

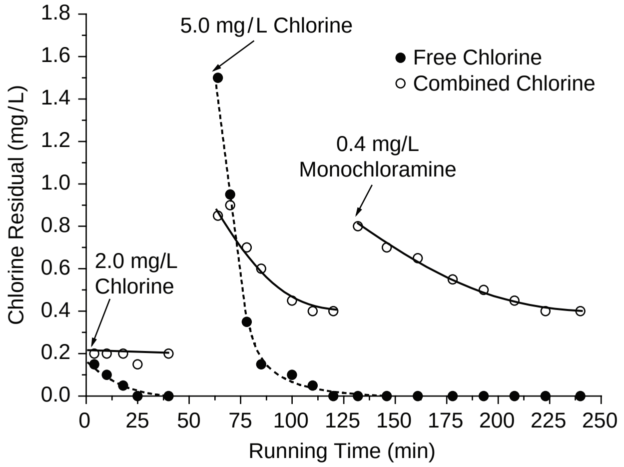

Fig. 3. Decline of chlorine residuals with time after both 2.0 and 5.0 mg/L chlorine dosing on the 83rd day of operation.

significant detachment as observed by the SS data in the carried flow during the course of chlorination (Fig. 2). Prior to chlorination, the SS level was 39 mg/L, but this was progressively reduced to 23 mg/L at the 10th min, and then to 9 mg/L at the end of 125th min.

Based on above findings, another chlorine dosing of 2.0 and 5.0 mg/L was followed on the next day (i.e., 83rd day). The test strategy on the 83rd day was to dose 2.0 mg/L first and let it run for 60 min; then another 5.0 mg/L was dosed again. The chlorine residual variations on the 83rd day are shown in Fig. 3. At the 120th min, the total residual was reduced to 0.40 mg/L, all of which was the combined residual. Because of this observation, another 0.40 mg/L mono-chloramine was added at the third hour of operation to raise the total combined residual to 0.80 mg/L. At the end of the 208th min, the total residual was reduced to 0.45 mg/L. It is to be noted here that during the second-day chlorination, the biofilm was actually subjected to a high chlorine exposure which would not be expected in the future full-scale disinfection for saline sewage effluent.

The biofilm response after the 83rd day chlorination is shown in Fig. 4. Just prior to chlorination, the SS level in the pipe flow was 38 mg/L. However, this quickly dropped to 22 mg/L in 5 min, and further dropped to 17 mg/L in one hour. Upon the second chlorine dosing of 5.0 mg/L, the SS level was found to further decline to around 10 mg/L for the remaining 150 min, even with the extra mono-chloramine dosing. These data clearly indicate that the two consecutive chlorine dosing of 2.0 and 5.0 mg/L did not materially affect the biofilm growth and sloughing, no matter whether the residual was present as free or combined form. Otherwise, the SS level in the pipe loop exit would have been greatly increased. It is believed that most of the added chlorine is either quickly consumed by the demand or converted to chloramine. The biofilm has a good tolerance for chloramine. It might be mentioned that just before chlorination on the 82th day, the average biofilm thickness was 157 μm. Then after two days of chlorination, the biofilm thickness was reduced to 118 μm.

In order to better show a "no impact" phenomenon, a plot of dynamic changes of biofilm thickness with time is shown in Fig. 5. The biofilm thickness continued to decline with the feeding of secondary effluent. Just prior to the first chlorination on the 80th day, the observed biofilm thickness was 157 μm. After two days of chlorination, the average thickness was reduced to 118 μm. However, such a reduction might not necessarily be caused by chlorination since even before that, there was a continuous decline in the biofilm thickness as a result of the low organic strength in the secondary effluent (BOD in the effluent was less than 10 mg/L). Similar to the low impact on biofilm thickness, the biofilm density and dry weight were not much affected by chlorination (data not shown). Based on the data of Figs. 2 and 4,

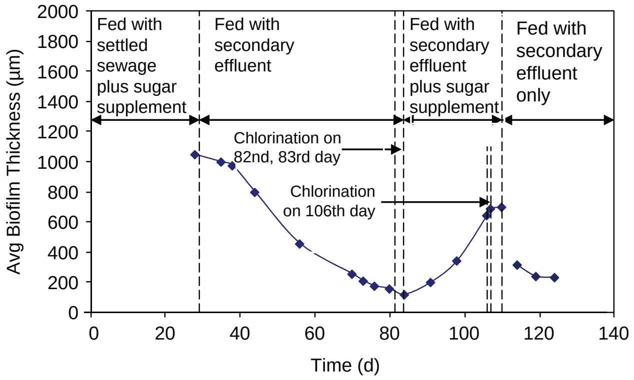

Fig. 5. Changes of biofilm thickness after feeding the secondary effluent.

chlorination of saline secondary effluent really did not induce any noticeable impact on the biofilm growth or sloughing.

To eliminate the possibility that the "no impact" observation was caused by low biofilm thickness prevailing at the time of testing, a thicker biofilm was allowed to build up after the 2.0 and 5.0 mg/L chlorination on the 83rd day. This was done by adding 100 mg/L sugar COD to the secondary effluent. The biofilm thickness was observed to quickly increase to 644 μm in three weeks. With such a thick biofilm, another round of chlorination at 5.0 mg/L was executed on the 106th day. To our surprise, there was still no adverse impact on the biofilm thickness, which continued to increase to over 700 μm in three more days (Fig. 5). Similarly, the biofilm density and its total dry weight were also found not much changed in the next several days after chlorine dosing (data not shown).

All of these apparently confirms our previous suggestion that chlorination of saline secondary effluent with a dosage of up to 5.0 mg/L does not affect biofilm growth or sloughing. This suggestion is consistent with the fact that in the water distribution system, there is always some biofilm growth although the water supply normally contains a free chlorine residual of 0.5–1.0 mg/L. In this study, the combined residual in the chlorinated effluent was only between 0.2 and 0.4 mg/L while the free residual was nil in most cases. Under such a situation, the chlorination impact on biofilm growth is not significant.

Finally, it is also of interest to note that on the 110th day of operation, the feed to the loop was switched to regular secondary effluent without any sugar supplement. With this change, the biofilm thickness and its total dry weight quickly declined to the previous low levels as observed before the first chlorination (on 82nd and 83rd day). From these data, it can be concluded that the existing biofilm growth in the 12-km transport system will not be much affected by the future chlorination operation.

3.2. Effect of sludge age on CBP formations

3.2.1. Cultivation of parallel activated sludge units with different sludge ages

To assess the impact of sludge age on both chlorine demand and the formations of THM and HAA, three activated sludge reactors were seeded with the mixed liquor suspended solids (MLSS) obtained from the actual sewage plant operation. The initial MLSS concentration in each reactor was 2,700 mg/L. Primary effluent from the plant was used for daily feeding. Every day the MLSS in each reactor was measured, and a calculated amount of MLSS was wasted in order to maintain a targeted sludge age. After the start, the biomass concentrations in all three reactors continued to decline with time. This was due to the low organic strength present in the primary saline effluent (around 20–30 mg/L DOC most of the time). The MLSS decline was faster with the shorter sludge age operation. After six weeks of cultivation, the MLSS level in all three reactors reached constant levels: 500–550 mg/L for 5-day sludge age, 700–750 mg/L for 8-day sludge age, and approximately 1,000 mg/L for 12-day sludge age. The final effluent DOC was approximately 10, 8, and 6 mg/L, respectively. The effluent from each reactor was then chlorinated with 2.0 and 5.0 mg/L dosing in order to assess the potential for the CBP formation.

3.2.2. CBPs formation

Table 1 shows the average TRC (total residual chlorine) and CBP formations after 60 min of contact at chlorination of 2.0 and 5.0 mg/L, respectively, to the effluents of the three different sludge ages.

At both chlorine dosing levels, the chlorine residual was slightly related to the sludge age. At 2.0 mg/L chlorination, mono-chloramine was the predominant residual in the treated effluent. When chlorine dosing was increased to 5.0 mg/L, some free residual appeared. A longer sludge age appeared to yield a lower TRC, which was attributed to a higher chlorine demand in the effluent. The lowest TRC of 0.28 and 0.90 mg/L, as found with 2.0 and 5.0 mg/L chlorination, were reasonably consistent with the values obtained from chlorinating the actual plant's effluent (i.e., 0.2 and 0.8 mg/L TRC at 2.0 and 5.0 mg/L chlorination) which was operated with a sludge age of 10–12 days.

Table 1 also indicates that a longer sludge age results in a greater specific yield of the total THM plus HAA on a per carbon basis. All of the CBP formation potentials are normalized to the DOC concentrations to obtain the specific yield, expressed as total CBP/DOC. In general,

Table 1
Chlorine residuals and CBP formations at different sludge ages.

		Chlorine residuals (mg/L as Cl_2)					
Sludge age (days)	Chlorine dosage	Free chlorine	Combined chlorine	TRC	Chlorine demands	DOC (mg/L)	CBP/DOC
5	2.0	0.05	0.38	0.43	1.57	10.85	1.07
8	2.0	0.00	0.30	0.30	1.70	8.25	2.24
12	2.0	0.03	0.25	0.28	1.72	6.79	4.68
5	5.0	0.55	0.90	1.45	3.55	10.85	1.58
8	5.0	0.40	0.53	0.93	4.07	8.25	5.84
12	5.0	0.45	0.45	0.90	4.10	6.79	7.24

a higher DOC in the effluent sample produced a higher absolute CBP; however, the differences in organic quality may also account for some reactivity difference on a molecular basis as reported by Ref. [17]. In Ref. [18] have also demonstrated that DOM in effluents from different sewage treatment plants may contain different kinds of organic matters. The data of this study seem to suggest that the remaining effluent organics from the longer sludge age operation have a higher reactivity with chlorine, and this results in a higher specific CBP/DOC yield.

Figure 6 illustrates the formation of different THM and HAA species at 5.0 mg/L chlorination of secondary effluents obtained from different sludge age operation (the 2.0 mg/L chlorination data are not shown here).

Similar to the above observation, an increase of sludge age from 5 to 12 days has also resulted in an increasing formation of both THM and HAA. However, in either case the formations of CBP are not high. To be more specific, at 2.0 mg/L chlorination, the productions of CBP are: 25 μg/L for THM and 7 μg/L for HAA_5. When chlorine dosing was increased to 5.0 mg/L, the formations of THM and HAA_5 are only slightly higher with a maximum level of 40 μg/L for THM and 10 μg/L for HAA_5 (Fig. 6). These CBP productions are well below the corresponding limits of 80 and 60 μg/L in the current USEPA (Year 2006) Drinking Water Standards [19].

Among the four THM species measured, $CHCl_3$, $CHCl_2Br$, $CHClBr_2$ are relatively low as compared to the major production of $CHBr_3$. Similarly, among the HAA formations, MCAA, MBAA are marginally detectable while DCAA and DBAA are the dominant species. Because the saline wastewater in Hong Kong is bromide-rich, the bromide ion may have shifted some CBP to bromine-containing species. In addition, bromine can also react with organic matter somewhat faster than chlorine. Due to the high bromide concentration

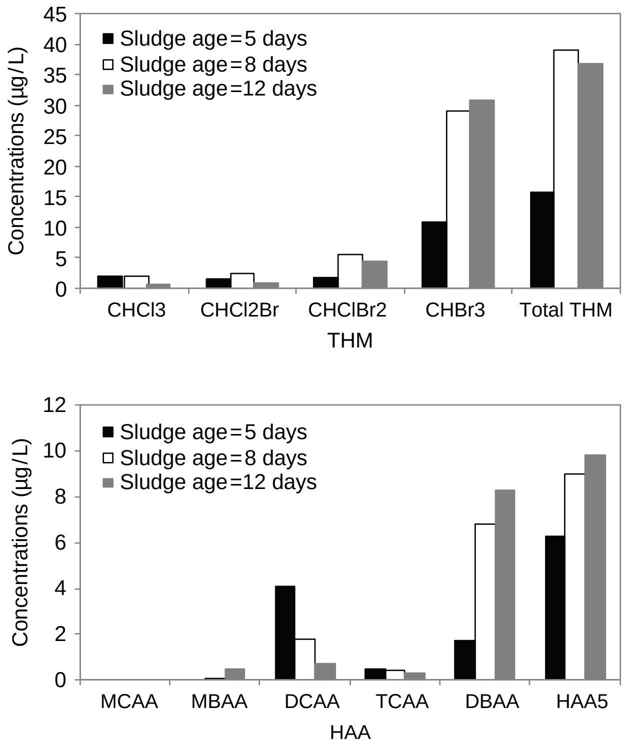

Fig. 6. Effect of sludge age on THM and HAA_5 formation at 5.0 mg/L chlorination and 60-min contact.

(around 22 mg/L) in the secondary effluent, brominated species, such as $CHBr_3$ and DBAA represent a large fraction of the total CBP. It is well known that the bromine-containing species generally has a greater public health concern than the chlorine counterpart [20]. Finally, it is also worth pointing out that in chlorinating the Hong Kong saline effluent from the activated sludge operation at 5, 8, and 12-day sludge age, the formation of total THM exceeds that of total HAA. This is in agreement with Yang's study [21] and the general trend of potable water chlorination [22].

4. Conclusions

The biofilm growth and sloughing are not affected by the presence of the remaining chlorine residual when the saline secondary effluent is disinfected by either 2.0 or 5.0 mg/L chlorination. As a consequence, the effluent SS level in the existing 12-km effluent transport system is not expected to increase after the full-scale chlorine disinfection is implemented.

Increasing sludge age from 5 to 12 days has resulted in a lower TRC, or a higher chlorine demand of the saline effluent. This has also resulted in an increasing formation of THM and HAA. Bromine-containing CBP are the dominant species owing to the bromide-rich saline wastewater in Hong Kong. However, in either case of 2.0 or 5.0 mg/L chlorination, the maximum formation levels of CBP are only 40 μg/L for THM and 10 μg/L for HAA_5, which are well within the current USEPA (Year 2006) Drinking Water Standards of 80 and 60 μg/L, respectively.

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