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Forecasting the chloramine residual in service reservoirs from online measurement

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ABSTRACT

In many systems, the occurrence of nitrification in chloraminated service reservoirs is the first sign of chloramine loss. Once the system is nitrified, recovery is difficult. To prevent the occurrence of nitrification, the risk has to be assessed well in advance. If chloramine residual, temperature and ammonia concentrations are predicted, the biostability concept could be used to assess the risk of nitrification. In this paper, the reservoir acceleration factor (F_{Ra}), which calculates chloramine decay rates within the reservoir from parameters measured online, has been used to forecast the most important parameter, chloramine residual. Results showed that F_{Ra} better shows the reservoir status than nitrite levels and the errors in the forecast residuals are less than 0.10 mg/l when predicted 10 d in advance. The distinct advantage of this approach is that it utilizes the online measurements as input variables such as temperature, inlet and outlet chlorine levels, hydraulic retention time obtained from reservoir level and flow meter readings. The approach opens an avenue to develop an online risk assessment tool, which will inform the utility of an imminent nitrification episode well in advance. The higher error than the measurement error (±0.03 mg/l) could be overcome by improving the model.

Keywords: Forecasting; Chloramine; Service reservoir; Nitrification; Online measurements

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1. Introduction

Chloramine is the second most popular disinfectant used in water distribution networks. In comparison to free chlorine, chloramine provides a better solution to maintain disinfectant residual in long distribution systems [1], produces less disinfection by-products and less secondary water quality issues (taste, odour and smell), due to less reactivity [2,3].

Despite advantages, maintenance of chloramine faces challenges at times, especially during summer. Microbial chloramine decay including nitrification is a major challenge to maintain residual chloramine level [4]. Nitrification is the microbiological oxidation of ammonia with oxygen into nitrite followed by the

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oxidation of nitrite into nitrate. The first step of nitrification is performed by ammonia-oxidizing bacteria (AOB) followed by nitrite-oxidizing bacteria (NOB). However, recent findings have shown that other species—e.g. *Nitrosomonas oligotropha* [5], nitrifying Archaea and heterotrophic nitrifiers [6]—can be present and contribute to nitrification. Therefore, in this paper, a common name ammonia-oxidizing microbes (AOM) is used.

Bulk water chloramine decay is affected in various ways. In the front end of the system, mild nitrification takes place until the residual reaches a residual sufficiently low enough to cause severe nitrification [7]. Mild nitrification is characterized by lower chloramine decay rates (0.001–0.004 h^{-1}), and lower but steady nitrite levels (<0.010 mg-N/L); severe nitrification is characterized by an order higher decay rates (~0.01 h^{-1}) and higher nitrite production rates resulting in higher nitrite concentration (>0.025 mg-N/L). If the nitrification status in the reservoir changes the microbial community [8] and decay characteristics [9] changes substantially.

In order to control the chloramine loss several short-term operational measures are practised, which include (1) reducing the water retention time in distribution systems [10,11]; (2) dosing free chlorine to increase the total chlorine to ammonia ratio thus diminishing the free ammonia residual [12,13]; (3) regularly carrying out breakpoint chlorination [11,12], (4) draining and refilling the service reservoir (tank) or diluting the nitrified water with freshly chloraminated water in winter [11,14] and (5) frequently flushing the distribution system [12].

Once nitrification commences, controlling the nitrification process is very difficult even by introducing the high chloramine dose through re-chloramination [15]. It is widely reported that breakpoint chlorination carried out after nitrification is not a desirable outcome as this can potentially introduce unwanted disinfection by-products and/or the process requires substantial resources [16]. In addition, letting nitrification to severe level before the decision to breakpoint chlorinate means customers are exposed to lower chloramine residual and undesirable disinfection by-products. Hence, it is wise to avoid the occurrence of nitrification.

Water spends majority of the time in reservoirs and reservoir holds majority of water making it a critical control point. In reservoirs, many factors contribute to the loss of chloramine such as water temperature, water retention time, mixing regime, stratification, biofilm bacterial activity, bulk water bacterial activity and the presence of sediments [9]. Most of these factors change with operational and environmental conditions and controlling nitrification is a challenge to the utility managers.

Reservoir nitrification is not a sudden process. Indicators could be used to understand the impending nitrification problem. However, the traditional indicators (chloramine, nitrite, nitrate, chlorine to ammonia ratio, ammonia, temperature, etc.) alone do not help in identifying the issue. Past studies have reported that the potential for nitrification exists when the chloramine concentration falls below the biostable residual concentration (BRC) [17,18]. As the chloramine residual reaches closer to the BRC, potential for nitrification or the risk of nitrification increases. Hence, the risk assessment can be based on the differences between the BRC and the predicted residual.

The BRC is a function of residual [7], temperature [17], inhibitor concentrations [18] and free ammonia concentration [7]. If temperature can be predicted, total ammonia concentration could be estimated and inhibitor is not present, total chlorine residual (assumed to be equal to the monochloramine residual, as it is the case under normal operating conditions at pH 8 and chlorine to ammonia molar ratio below 1:1) could be used to estimate the free ammonia concentration, and hence, the BRC could be established and the future risk could be estimated.

To forecast chloramine residual, chloramine decay rate in the reservoir should be known. The chloramine decay present in the reservoir can be understood from both reservoir acceleration factor (F_{Ra}) and F_{m} concept [4,9]. The F_{m} method quantifies the roles of microbes and chemical agents in decaying chloramine in bulk waters of the reservoir from laboratory measurement. The method measures chloramine decay rates in the original and microbial activity inhibited (or microbes removed by filtration) samples and names them as the total (k_t) and chemical decay (k_{c}) rate coefficients. The difference between k_t and k_{c} is referred to as the microbial decay rate coefficient (k_{m}). The F_{m} is the ratio between the microbial decay coefficient (k_{m}) and the chemical decay coefficient (k_{c}) (Eq. (1)).

$$F_{\mathrm{m}} = \frac{k_{\mathrm{m}}}{k_{\mathrm{c}}} \tag{1}$$

where k_{m} and k_{c} are the microbial and chemical decay coefficients, respectively.

The F_{Ra} concept, on the other hand, calculates chloramine decay rates in the whole reservoir content using online measurements and defines the degree of acceleration present in the reservoir over and above the chemical decay in the bulk water [9]. The chemical decay rate coefficient used is the base chemical decay rate coefficient which is the decay that would occur if no microbial influence is present in the sample. The best (lowest) chemical decay is usually experienced under mild nitrifying conditions and is relatively stable. As the operator wants to know how far away the chloramine decay behaviour in the reservoir compared to the best case scenario, Eq. (2) was suggested to calculate the reservoir acceleration factor, F_{Ra}.

$$F_{\mathrm{Ra}} = \frac{(k_{\mathrm{Rt}} - k_{\mathrm{bc,T}})}{k_{\mathrm{bc,T}}} \tag{2}$$

where k_{Rt} is the chloramine decay coefficient (h^{-1}) within the reservoir and $k_{\mathrm{bc,T}}$ is the adjusted base chemical decay coefficient (h^{-1}), at the reservoir water temperature (T in ℃) and k_{Rt} is estimated at the reservoir water temperature from online measurements assuming the reservoir contents are completely mixed as shown in Eq. (3).

$$k_{\mathrm{Rt}} = \frac{(TCl_{\mathrm{in}} - TCl_{\mathrm{out}})}{\theta \cdot TCl_{\mathrm{out}}} \tag{3}$$

where TCl_{out} and TCl_{in} are the outlet and inlet chloramine residuals (as total chlorine), respectively, and θ is the retention time in the reservoir.

The $k_{\mathrm{bc,T}}$ is the converted decay coefficient from the base chemical decay coefficient to the reservoir water temperature using Eq. (4).

$$k_{\mathrm{bc,T}} = k_{\mathrm{bc,20}}\ \exp\left[-\frac{E}{R}\left(\frac{1}{(273+T)} - \frac{1}{(273+20)}\right)\right] \tag{4}$$

where E/R value is estimated to be 3,551 K^{-1} [19] and $k_{\mathrm{bc,20}}$ is the base chemical decay coefficient, which is estimated as the average chemical decay coefficient measured (using the F_{m} method) in mild nitrifying bulk water samples of the water supplied to the reservoir in the preceding three months.

In summary, operators need to understand whether a reservoir will undergo nitrification now and in the future given the temperature and operational conditions could be predicted. Forecasting chloramine residual is critical to understand the risk of nitrification. There is no reported method to forecast such using online measurements. Therefore, the main objective of this paper is to develop a method to forecast future chlorine level in the reservoir from online measurements. Considering the data availability and operational conditions, chlorine level is predicted 10–20 d in advance. The method is developed first and then validated with a full-scale reservoir data set (2003–2005) obtained from Sydney Water Corporation.

1.1. Method development

The total decay rate coefficient within the reservoir is estimated as k_{Rt} in the F_{Ra} method at water temperature using Eq. (3). If the decay rate coefficient in the future (at another temperature) is needed, two main factors are to be considered: change in chemical and microbial components of k_{Rt} and change in microbial community.

If chemical and microbial components of k_{Rt} can be separated, Eq. (4) can be used with different values for E/R to represent chemical and microbial components [19]. Sathasivan et al. showed E/R values for chemical and microbial chloramine decay as 3,551 and 6,924 K^{-1}, implying that the microbial decay coefficient changes more than that of chemical decay [19]. The operators want to know the conservative estimate of the residual, i.e. the lowest possible residual. Hence, rather than separating chemical and microbial decay coefficient, the whole decay is assumed to behave like microbial decay coefficients.

During the forecast time, microbial community changes in many different ways. First, the growth may occur due to the increase in temperature or the drop in residual. Similarly, number of microbes may decrease, for example during autumn, decreasing the decay coefficient. Microbial community may change due to the change in conditions such as the onset of nitrification introducing drastic changes to microbial decay coefficient [7,8]. Taking this into account needs a proper study and will lead to a better prediction over a very long time. However, if a shorter time frame is used and the decay coefficient in the reservoir is conservatively assumed to behave like microbial decay coefficient with an E/R value of 6,924 K^{-1} [19] and the prediction before the onset of nitrification is what needed, then microbial community changes could be neglected. Hence, the k_{Rt} value in any future temperature T_1 can be estimated using Eq. (5).

$$\frac{\mathrm{k}_{\mathrm{Rt(pred)}}}{k_{\mathrm{Rt}}} = \exp\left[-\frac{E}{R}\left(\frac{1}{(273+T_1)} - \frac{1}{(273+T)}\right)\right] \tag{5}$$

where k_{Rt} is the latest decay coefficient (h^{-1}) calculated at the time of forecasting and $k_{\mathrm{Rt(pred)}}$ is the predicted decay coefficient (h^{-1}) after a nominated lapsed time, T is the latest water temperature (℃) measured at the time of forecasting; T_1 is the expected temperature (℃), at which the residual needs to be predicted. E/R value in Eq. (5) is 6,924 K^{-1}.

The reservoir chloramine level can be predicted with Eq. (6).

$$TCl_{\text{out,pred}} = \frac{TCl_{\text{in,exp}}}{1 + \theta k_{\text{Rt(pred)}}} \tag{6}$$

where $TCl_{\text{out,pred}}$ is the predicted outlet chloramine residual, $TCl_{\text{in,exp}}$ is the expected inlet chloramine residual (as total chlorine) and θ is the retention time in the reservoir. Assuming the operation and maintenance plan for the next few days is fixed, the retention time of the reservoir can be predicted.

In Sydney Water systems, water temperature fluctuates between 12 and 24℃ (within six months, it raises and in the next six months, it drops back). Therefore, the average temperature increase and drop are around 0.67℃ every 10 d. In this study, future temperature is conservatively assumed to increase by 1℃ every 10 d from September to February and decrease by 0.5℃ from March to August. The future retention time and inlet chloramine residual are assumed to be the same as the current one. Microbial growth inside the reservoir is neglected within the forecast period.

2. Materials and methods

A full-scale reservoir data were used to validate the approach. This reservoir is located in one of the Sydney's main distribution subsystems. The reservoir is an elevated circular tank of diameter 18 m, height 13 m and capacity of 3×10^3 m^3. As the reservoir is situated in a bushfire prone area, the highest possible water level must be maintained during summer. The upstream reservoir does not experience nitrification. Retention time in the reservoir during the sampling period was 2.4–2.9 d. The reservoir has a common inlet/outlet pipe of 0.45 m diameter, opening 1.0 m above the bottom and at the reservoir centre. Its surface area to volume (S/V) ratio was 0.31 m^{-1}. Mixing in this reservoir is achieved only by inflow.

The data set, generated from 2004 to 2005, consisted of various parameters necessary for the model: reservoir retention time, inlet total chlorine levels and outlet temperature and concentrations of total chlorine, ammonia, nitrite and nitrate.

NO_2-N, NO_3-N and total NH_3-N concentrations were measured using the flow injection analysis method [20]. NO_2-N was measured by the sulphanilamide method [20]. NO_3-N was first reduced by the cadmium reduction method to NO_2-N and then NO_2-N was measured by the sulphanilamide method [20]. NO_2-N had the lowest detection level of 0.002 mg/L. NH_3-N was measured by the phenate method [20]. Total chlorine residuals were measured by the DPD colorimetric method using a HACH pocket colorimeter [20]. Total chlorine measurement had an experimental error of 0.03 mg/L.

Even though the data set was complete, the sampling interval was not consistent and varied from 7 to 30 d. In order to validate the model, reservoir data in every 10 d were required and missing data were linearly interpolated.

3. Results and discussion

3.1. General behaviour of the reservoir

Figs. 1A and 1B show the behaviour of reservoir A during the two years of observation. Average inlet chlorine level of the reservoir was recorded as 1.4 mg/l (1.3–1.6 mg/l) and the retention time was around 2.5 d (Fig. 1A). Water temperature in the summer seasons had exceeded 23℃ and in winter seasons,

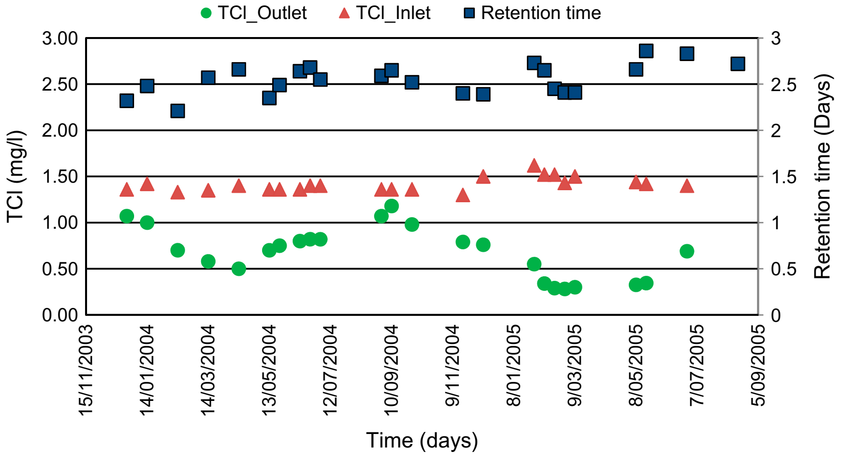

Fig. 1A. Inlet and outlet total chlorine levels and the retention time of the reservoir.

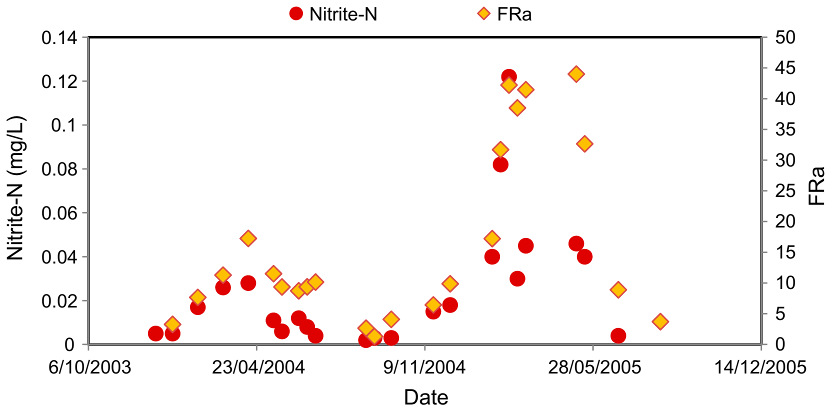

Fig. 1B. Variation of nitrite and F_{Ra} in the reservoir.

it had dropped to 12°C. Calculated F_{Ra} levels show that reservoir content had higher (>3) values even in winter (Fig. 1B), showing the reservoir contains microbes sufficient to decay chloramine more than three times the chemical decay. In summer 2004, the F_{Ra} was even higher (~17) at the time when nitrite levels reached only 0.028 mg/L which usually begins to show some concern.

More interestingly, the F_{Ra} values had similar trend to nitrite, but showed that F_{Ra} can be a better indicator because even when utilities feel safe in terms of nitrite (<0.025 mg-N/L) and chloramine levels (>0.8 mg/L), chloramine had been decaying to a much higher degree. F_{Ra} can be calculated from online measurement and it shows what happens to the chloramine residual. Hence, F_{Ra} is a better parameter for monitoring a chloraminated reservoir.

It is observed that the reservoir nitrite (NO_2-N) levels have exceeded 0.125 mg/l during summer, suggesting nitrification episodes. It is desirable if the outlet chloramine residuals (thus the risk of nitrification using the biostability concept) can be predicted before the onset of nitrification. As chloramine is the primary parameter for this needs to be predicted.

3.2. Method validation, statistical analysis

Figs. 2A and 2B summarize the actual outlet *TCl* levels, predicted *TCl* levels (10 and 20 d in advance), nitrite and error in each sampling interval. In general, *TCl* levels predicted 10 d in advance are more accurate than those predicted 20 d in advance. Larger errors have occurred when nitrite levels have started to increase (shown within the rectangles) and when

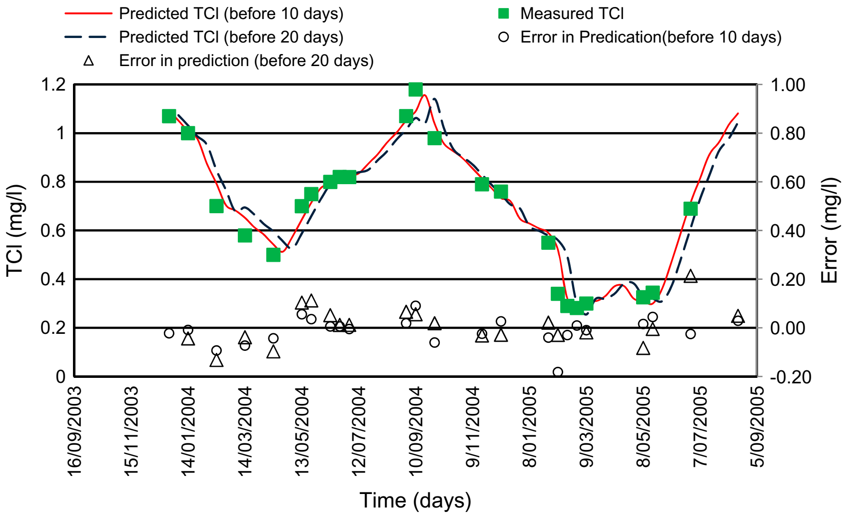

Fig. 2A. Variation of predicted *TCl* level (10 and 20 d), actual *TCl* level and the error.

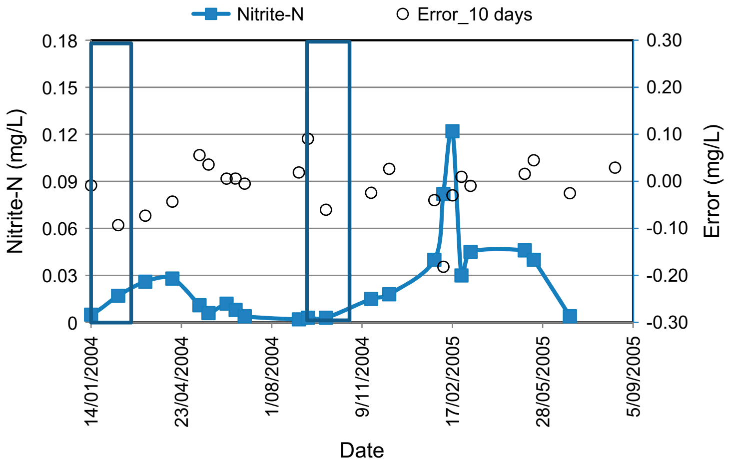

Fig. 2B. Variation of nitrite nitrogen level and error for predicted *TCl* level (10 d in advance) with time.

cnitrification was at severe levels that required intervention (as noted within the peak of nitrite level on 17 Feb 2005). In the method development, it was assumed that the microbial community and retention time in the reservoir remain the same. The model was working well over a long period of time except after the onset of nitrification. The purpose of the method is to forecast chloramine residual before the onset of nitrification, and hence, the model has performed well except for one instance on 10 Sept 2004.

3.3. Application and limitation of the method

In large water distribution networks, a number of reservoirs with different characteristics (size, shape and location) are connected by a complicated pipe network. Outflow of one reservoir may be the inflow of another reservoir. Complex situation in water distribution networks makes modelling of chloramine level in the network extremely difficult. However, the proposed method opens an avenue to forecast the chloramine level in the distribution network. The method can be used to calculate chloramine loss in each reservoir and the combined analysis with hydraulic properties of the network (connection among the reservoirs) will give a promising approach to forecast chloramine level in any place in the distribution network.

Future chloramine level in the reservoir is predicted assuming several future operational parameters that are known. Some of these parameters may be difficult to forecast (e.g. reservoir level, water temperature and inlet chloramine level).

For the calculations to be valid, the microbial community in the reservoir should not change very much. It is well known that the significant microbial community changes occur at the onset of nitrification and hence the method becomes invalid after the onset (7, 8). When chloramine residuals are predicted, the BRC concept should be applied to understand the point of onset and thus the invalid region of the method. Because once the predicted residual falls below the BRC, the predication will not be valid.

For the F_{Ra} concept to provide accurate reflection of the reservoir contents, it should have a completely mixed condition. However, reservoir stratification ensures a longer retention time for the near-surface water and enhances the chemical and microbial chloramine decay, thereby, decreasing the chloramine residuals substantially in surface layers. Ike et al. showed that, in summer, AOB concentrations are 10–20 times higher in the surface layer (0.3 m below surface) compared with 5 m below the surface [10]. In addition, previous studies have reported the presence of microbial stratification during summer and winter in unmixed reservoirs despite chemical and thermal homogeneity [21]. The status of the reservoir in terms of mixing can be found by applying F_{Ra} concept and F_{m} method simultaneously as demonstrated elsewhere [9].

4. Conclusion

The reservoir acceleration factor (F_{Ra}) method is useful in providing online measurement of status of a reservoir better than nitrite levels. The proposed method had an experimental error less than 0.10 mg/L compared to the measured residuals without the consideration of the impact of microbial community changes on chloramine decay rates which is the case before the onset of nitrification. Therefore, this tool is effective to forecast chloramine levels in water distribution network when it is mostly needed, i.e.

before the onset of nitrification. The method could also be used in analysing various operational scenarios but care should be taken to neglect any predicted residual below the BRC since nitrification would onset below the BRC. The approach is simple as it requires easily measurable parameters of inlet chlorine level, reservoir water temperature and reservoir retention time. Most importantly, because the method relies on parameters that can be measured online, it also opens an avenue to warn of possible reservoir nitrification risk using online measurement.

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Nomenclature

AOB — ammonia-oxidizing bacteria
AOM — ammonia-oxidizing microorganisms
NOB — nitrite-oxidizing bacteria
F_{Ra} — reservoir acceleration factor (–)
BRC — biostable residual concentration (mg-$\mathrm{Cl_2}$ $\mathrm{L^{-1}}$)
F_{m} — microbial decay factor (–)
k_{m} — microbial decay coefficient in bulk waters as measured during microbial decay factor method measurement ($\mathrm{h^{-1}}$)
k_{c} — chemical decay coefficient in bulk waters ($\mathrm{h^{-1}}$)
k_{Rt} — the total decay of chloramine present in the reservoir at the reservoir water temperature ($\mathrm{h^{-1}}$)
k_{bc} — base chemical decay coefficient at 20°C (average of current chemical decay coefficients (k_{c}) measured in the past one–three months) ($\mathrm{h^{-1}}$)
$k_{\mathrm{bc,T}}$ — base chemical decay coefficient converted to the reservoir water temperature, T (°C) ($\mathrm{h^{-1}}$)
TCl_{in} — current inlet chlorine residual (mg-$\mathrm{Cl_2}$ $\mathrm{L^{-1}}$)
TCl_{out} — current outlet chlorine residual (mg-$\mathrm{Cl_2}$ $\mathrm{L^{-1}}$)
θ — current retention time of the reservoir (h)

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