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Chlorine decay in water supply pipelines: impact of nitrogen concentration in the source waters

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ABSTRACT

Chlorine decay in two water mains in the Metropolitan Region of Fortaleza, Brazil was measured and modeled. Consistent bulk and wall decay constants were obtained. The time-variations of chlorine decay suggest that nitrogen concentration in the source waters has a significant impact on chlorine consumption. This has been attributed to potential reactions with nitrogenous material. Correlations were proposed for prediction of the bulk and wall decay constants as functions of the nitrogen concentration and relative roughness of the pipes, respectively. Model simulations better fitted the experimental data than assuming constant decay. Moreover, instead of using an approximately constant chlorine concentration at the water treatment plant outlet, as currently done by the water utility company, the simulations revealed that the management goal of 0.2 mg/L can be met at network extremities, without the need for rechlorination along the system, by setting optimum values of chlorine as a function of nitrogen concentration.

Keywords: Chlorine consumption; Modeling; Water distribution; Water pollution

1. Introduction

The maintenance of adequate water quality conditions in water distribution systems (WDS) is crucial for human health. In this sense, chlorine has long been used for disinfection purposes in WDS. Thus, a free chlorine residual must be maintained in WDS in order to reduce the risk of pathogen contamination. However, chlorine concentration decreases with time and space due to consumption. This temporal and spatial consumption is caused by chemical reactions of chlorine with water constituents and the pipe wall [1–7]. Therefore, understanding and controlling these chemical reactions is important to optimize the operation of WDS regarding water quality.

Many studies investigated the chlorine decay in WDS. The authors usually consider the plug-flow approach and first-order reaction, assuming two chlorine decay constants: a bulk reaction coefficient (k_b) and a wall reaction coefficient (k_w) [8–13]. Complete mixing at pipe junctions is also normally assumed. The bulk reaction coefficient has been related to many factors such as water temperature, initial chlorine concentration, dissolved organic matter and inorganic substances, such as ammonia, iron and manganese [14–17]. On the other hand, the wall reaction coefficient has

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been described as a function of pipe diameter, material and age, but the reactions with the biofilm formed at the pipe wall and corrosion particles have also been related to the above-mentioned water quality parameters [18–21].

The values of k_b and k_w are determined from experiments, model calibration and/or predictive equations [8,14,16,17,22,23]. Hence, the aforementioned formulations (plug-flow approach considering first-order reaction and complete mixing at junctions), together with predefined values of k_b and k_w, have been frequently used in computational packages such as EPANET for hydraulic and water quality modeling of WDS [24]. More complex models, including other reaction schemes and the reaction and transport of multiple species such as dissolved/suspended constituents and chemicals adsorbed onto particles, are also available in the literature [2,15,25] and in commercial softwares such as EPANET-MSX, which is an extension to the original EPANET [26]. Incomplete mixing at pipe junctions has also been considered both experimentally and computationally [27,28].

In Brazil, the Ministry of Health issued Ordinance #2914/2011 establishing a mandatory range of 0.2–2.0 mg/L for free chlorine residual in WDS. Therefore, it is usual to add a significant dose rate of chlorine at the water treatment plants (WTP), so that the upper limit of 2.0 mg/L must be established at the entry to the WDS, while the management goal of 0.2 mg/L must be achieved at network extremities, without the need for rechlorination along the system. However, chlorine concentrations much higher than 2.0 mg/L can generate high water treatment costs and also result in taste/odour problems, potentially promoting chemical reactions with water components that produce harmful substances to human health such as trihalomethanes (THMs) [29,30]. Hence, it is desirable to optimize the system to reach these limits and avoid the above-mentioned issues. On the other hand, to meet this goal, it is necessary to obtain accurate values of k_b and k_w to provide a precise simulation of chlorine concentration throughout the water supply systems. Nevertheless, only a few studies in Brazil performed calibration of these constants, resulting in wide ranges of values of k_b (0–5 d^{-1}) and k_w (0–0.6 m/d), which were fitted from simulations of hypothetical water distribution networks [31–33]. Although these values are within the ranges reported in the classical literature [1,8,9], because of anthropic pressure, water supply reservoirs in Brazil normally present high levels of nitrogen and related compounds such as ammonia, which are not significantly removed in conventional WTPs. This potentially has an impact on chlorine decay in water transmission and distribution systems, as disinfection efficiency is expected to reduce due to chlorine reaction with ammonia [34], if the drinking-water contains more than 0.2 mg/L of this compound [35].

In this study, we carried out measurements of chlorine concentration upstream and downstream in two water mains of different diameters, materials and flow conditions in the Metropolitan Region of Fortaleza (MRF), Brazil, in order to investigate the chlorine decay along these pipelines. The systems connect a conventional WTP fed by a hypereutrophic reservoir to two municipalities. The bulk and wall decay constants (k_b and k_w) were fitted in EPANET by using chlorine concentration data collected every 2 h for a total duration period of 32 months. The relationships between these decay constants, pipe characteristics and nutrient concentrations in the water supply reservoir were also investigated. Finally, new correlations were proposed to improve the prediction of chlorine consumption in water supply pipelines in Brazil. EPANET simulations were also provided to illustrate the applicability of the results for the optimization of the operation of water treatment, transmission and distribution systems regarding chlorine disinfection.

2. Methodology

The studied system consists of two drinking water transmission pipelines with flow rates Q of 60 and 500 L/s that supply the city of Caucaia and a neighborhood of Fortaleza called Pici, respectively, in the Metropolitan Region of Fortaleza, State of Ceará, Brazilian northeast. As shown schematically in Fig. 1, these pipelines are made of steel and cast iron and have lengths L of 6.2 and 15 km and diameters D of 400 and 1,500 mm, respectively. A conventional WTP with capacity of 1.2 m^3/s supply both pipelines. This WTP is fed by a hypereutrophic surface water reservoir called Gavião, with capacity of 33 hm^3. The water treatment consists of coagulation, flocculation, sedimentation, followed by double sand-filtration and final disinfection with gaseous chlorine. As reported by the Water and Sewage Company of Ceará (CAGECE), average chlorine concentration (C) is 2.70 mg/L at the WTP outlet and about 2.0 mg/L at the inlet of Caucaia and Pici water distribution networks. On the other hand, the raw and treated waters have relatively high ammonia concentrations of 0.1–0.5 mg/L, while nitrate and nitrite concentrations are usually lower than 0.1 mg/L or below the detection limit. Similar ammonia concentrations are also observed in other Brazilian surface water reservoirs and WTP's, which suggest that a chlorine demand along the transmission pipelines is expected [35].

Data of chlorine concentration measured with a chlorine analyzer with range of 0–5 mg/L (DIGIMED AI-CL2-HP) was provided by CAGECE. Chlorine concentration was measured every 2 h for a total duration of 32 months (Jan 2018–August 2020) at the outlet of the WTP and inlet of the water distribution networks of Caucaia and Pici (Fig. 1). Note that smooth changes of chlorine concentration were observed considering the 2 h intervals and the total variation of this parameter within each month was relatively small (up to about 30%). Additionally, the measurements followed approximately normal distributions with mean and median values varying within ±5%. Thus, using monthly averaged values of chlorine concentration, the bulk and wall decay constants (k_b and k_w) for each pair of upstream and downstream chlorine concentrations were fitted in EPANET, according to the following procedure:

- A wall reaction formula was used for both water mains: $k_w = -F/[\log(\varepsilon/D)]$ [24], 2020), in which F is a fitting parameter, while relative roughness (ε/D) values of 2.5E-03 and 6.7E-05 were used for the cast iron (WTP-Caucaia) and steel (WTP-Pici) pipelines, respectively.

- A single value of F for both pipelines and monthly values of k_b for each pipeline were adjusted by fitting EPANET model simulations to measured data for each particular month. This included a one-step minimization of the following objective function, in which the variables F and k_b were fitted by using the principle of least squares:

$$\underset{F,k_b}{\text{Minimize}}: \sum_{i=1}^{n}\left[C_i\left(F,k_{b,i}\right)-C_{m,i}\right]^2 \quad (1)$$

where $C_i(F,k_{b,i})$ = chlorine concentration predicted by the model at the downstream sections of both pipelines (Caucaia and Pici); $C_{m,i}$ = chlorine concentration measured at the downstream sections of both pipelines; and n = number of experimental data points (64 in total, including the data of both pipelines).

The values of k_b obtained for each month and pipeline were compared with nutrient concentrations in the source waters (Gavião Reservoir), which were provided by the Water Resources Management Company of Ceará (COGERH). Note that the monitoring program includes measurements of total nitrogen (N) and total phosphorus (P) concentrations at the water intake of 155 surface water reservoirs monitored by COGERH, with capacities ranging from about 1–6,700 hm^3. Similar monitoring programs focusing on N and P are also conducted in other water supply reservoirs in Brazil, as reported by the National Water Agency (ANA). Observe that, since the Gavião Reservoir was hypereutrophic, both N and P components such as ammonia, organic nitrogen and orthophosphate concentrations are expected to be relevant in the source waters. However, as detailed data regarding the fractions of nitrogen and phosphorus were not available, we decided to use total nitrogen and total phosphorus in our analysis.

Additional data of water temperature and biochemical oxygen demand (BOD) of the raw and treated waters, as well as flow rate and pressure at the monitoring points were also provided by CAGECE.

After analysis of the data, the combination of chlorine and N concentrations allowed the fitting of a new correlation to predict the bulk decay constant k_b. Hence, a comparison of the predictability of the EPANET model by using average values of k_b and the values obtained from the proposed correlation [$k_b = f(N)$] was also performed for both pipelines.

Finally, the proposed correlations for both k_w and k_b were used to simulate different scenarios of chlorine concentration in the water distribution network of Caucaia, containing PVC and cast iron pipelines with diameters ranging from 50 to 400 mm. The idea was to optimize the operation of the WDS regarding chlorine disinfection.

3. Results and discussion

Fig. 2 shows time-series of chlorine concentrations (C) measured at the upstream (at WTP) and downstream (at Caucaia and Pici) transects of the two drinking water transmission pipelines, as well as total nitrogen (N) and total phosphorus (P) concentrations measured at the source waters (Gavião Reservoir). In the upper figure, it is clearly seen that while C measured at the WTP present a maximum variation of about 7%, the values measured at Caucaia and Pici present much more pronounced variations of 47% and 65%, respectively. Since the variations of water temperature, BOD, flow rate and pressure at the monitoring points were all lower than 15%, it can be inferred that the large variations in C at Caucaia and Pici sections may be related with the variations of nutrient concentrations at the source waters, which corresponded to about 82% and 94% for N and P, respectively. However, while high N values were in general related to high chlorine decay, P did not present any clear trend with chlorine concentration in the pipelines. Thus, the effect of N on C decay may be attributed to additional chlorine demand due to potential reactions with nitrogenous material such as ammonia and organic nitrogen [34,35].

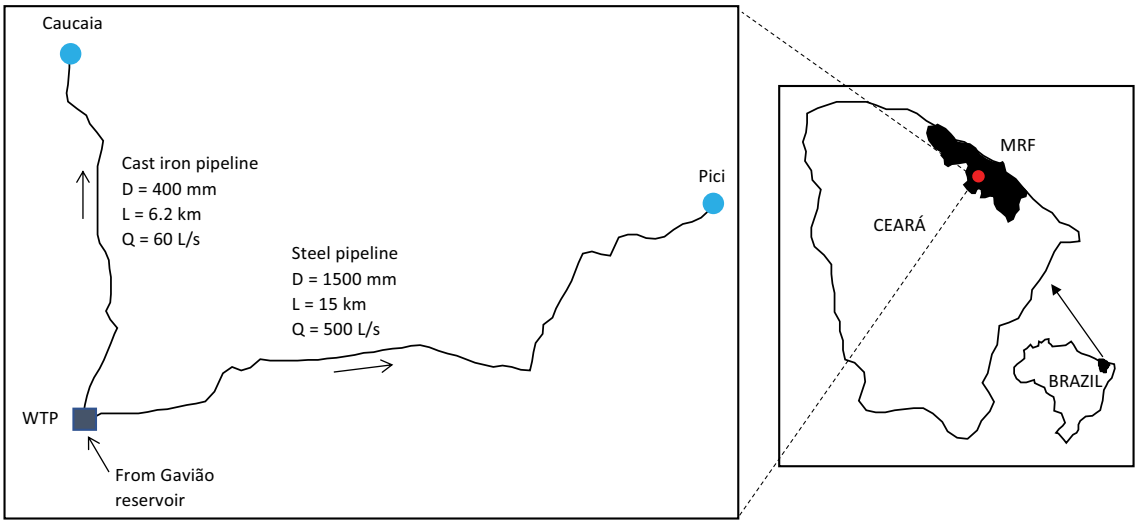

Fig. 1. Schematic of the two drinking water transmission pipelines investigated in the present study. MRF stands for Metropolitan Region of Fortaleza, Brazil.

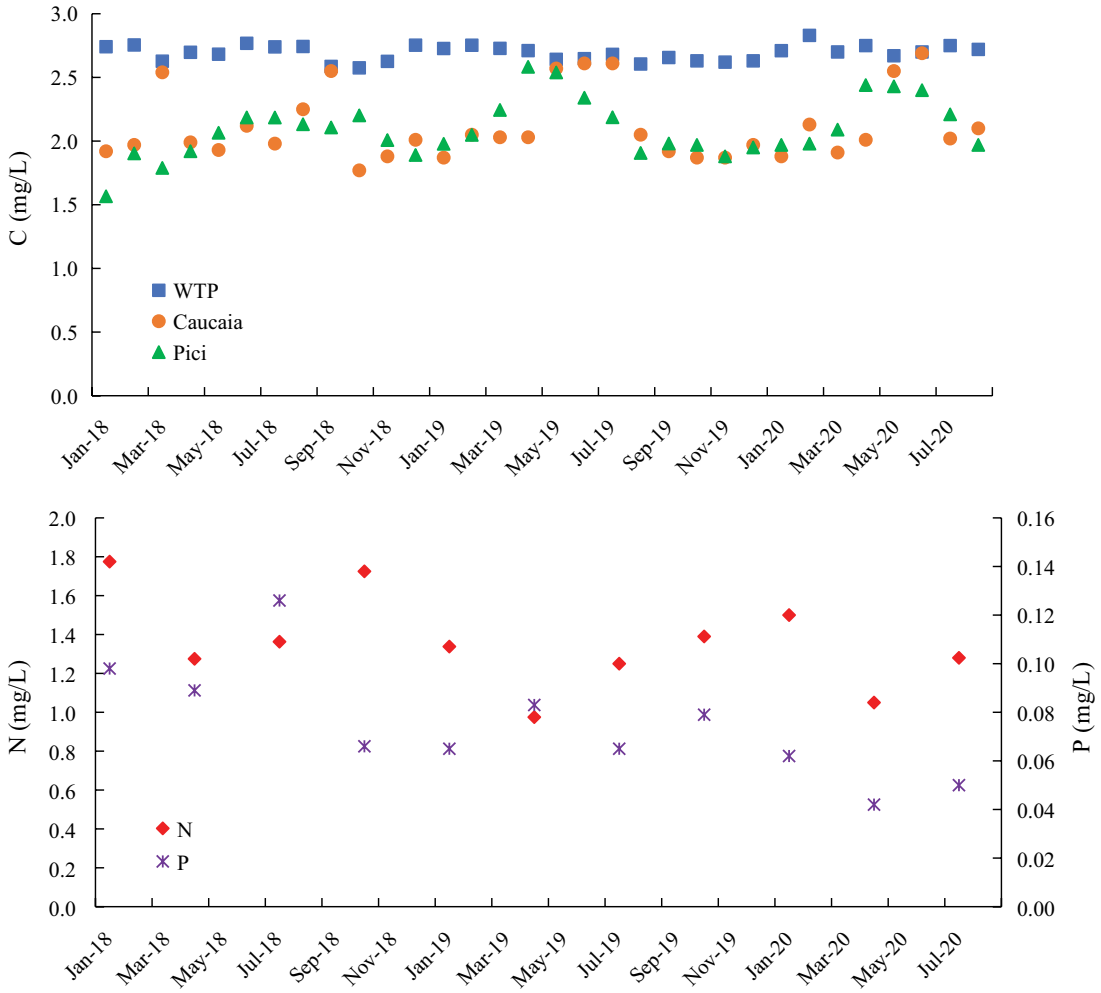

Fig. 2. Time-series of chlorine concentrations (*C*) measured at the upstream (at WTP) and downstream (at Pici and Caucaia) transects of the two drinking water transmission pipelines, and nitrogen (*N*) and phosphorus (*P*) concentrations measured at the source waters (Gavião Reservoir).

The minimization of the sum of the squares given by Eq. (1) resulted in standard deviations between EPANET model simulations and measurements lower than 6%, as well as in values of k_b ranging from about 0–0.8 d^{-1} (average of 0.2 d^{-1}) and F = 0.5 m/d, corresponding to k_w = 0.2 and 0.1 m/d [$k_w = -0.5/\log(\varepsilon/D)$] for the water supply pipelines of Caucaia and Pici, respectively. The fitted values of k_b and k_w are within the ranges reported in the literature for similar flow conditions, pipe dimensions and materials [1,8,9,31–33]. This implies that these values can be used to represent the general behavior of chlorine decay in both pipelines. The assumption of constant k_w for approximately constant initial chlorine concentration ($C \approx 2.70$ mg/L) and water temperature ($T \approx 28$°C), as well as for fixed pipe diameters/materials, is consistent with Lee et al. [21].

Fig. 3 evaluates the effects of *N* and *P* concentrations on the behavior of chlorine decay. While a good correlation was obtained between k_b and *N* (R^2 = 0.7172), k_b did not correlate with *P* (R^2 = 0.0056), as expected. Note that due to the lower frequency of *N* and *P* measurements (every three months), as compared to *C* measurements, the data of k_b correspond to three-month averages. The linear increase of k_b with *N* corroborates the results of Vieira et al. [17], in which k_b was linearly related with other water quality parameters such as organic matter and iron content. This suggests that water pollution influences directly the consumption of chlorine in the pipelines. However, the results obtained in the present study imply that reactions with nitrogenous material such as ammonia, as pointed out by Zhou et al. [34] and WHO [35], are probably the cause of the faster chlorine decay when *N* concentration is higher. This may have an important impact on water supply systems in Brazil, notably in the northeast region, as *N* concentration in the surface water reservoirs is usually high due to deficient sanitation services and agricultural runoff [36–38].

Fig. 4 shows comparisons between modeled and measured chlorine concentrations for both pipelines (Caucaia and Pici), considering two decay conditions: (1) a constant value of k_b = 0.2 d^{-1} for both pipelines; and (2) a variable k_b as a function of nitrogen concentration

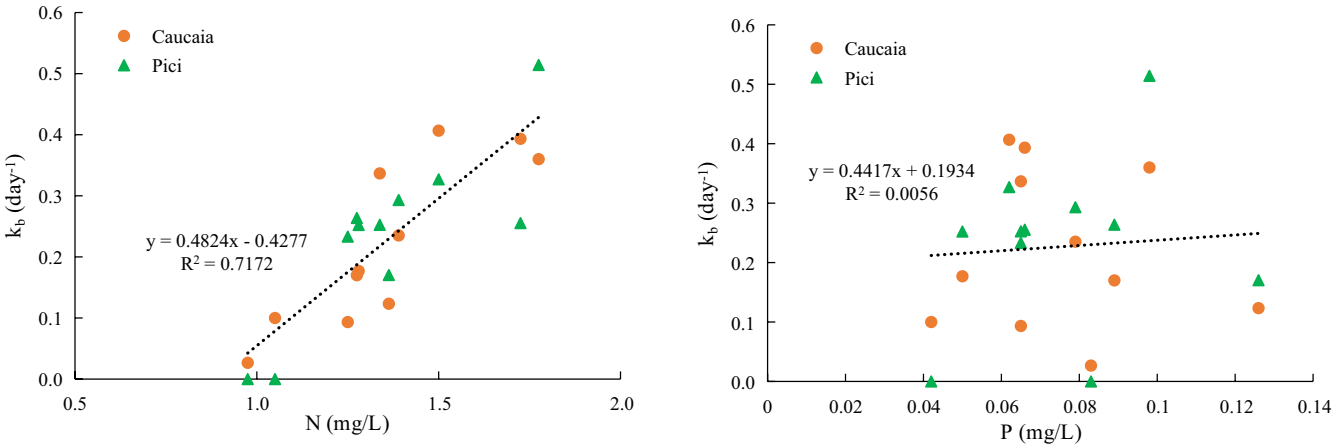

Fig. 3. Adjusted correlations for the bulk decay constant (k_b) as functions of nitrogen (N) and phosphorus (P) concentrations at the source waters.

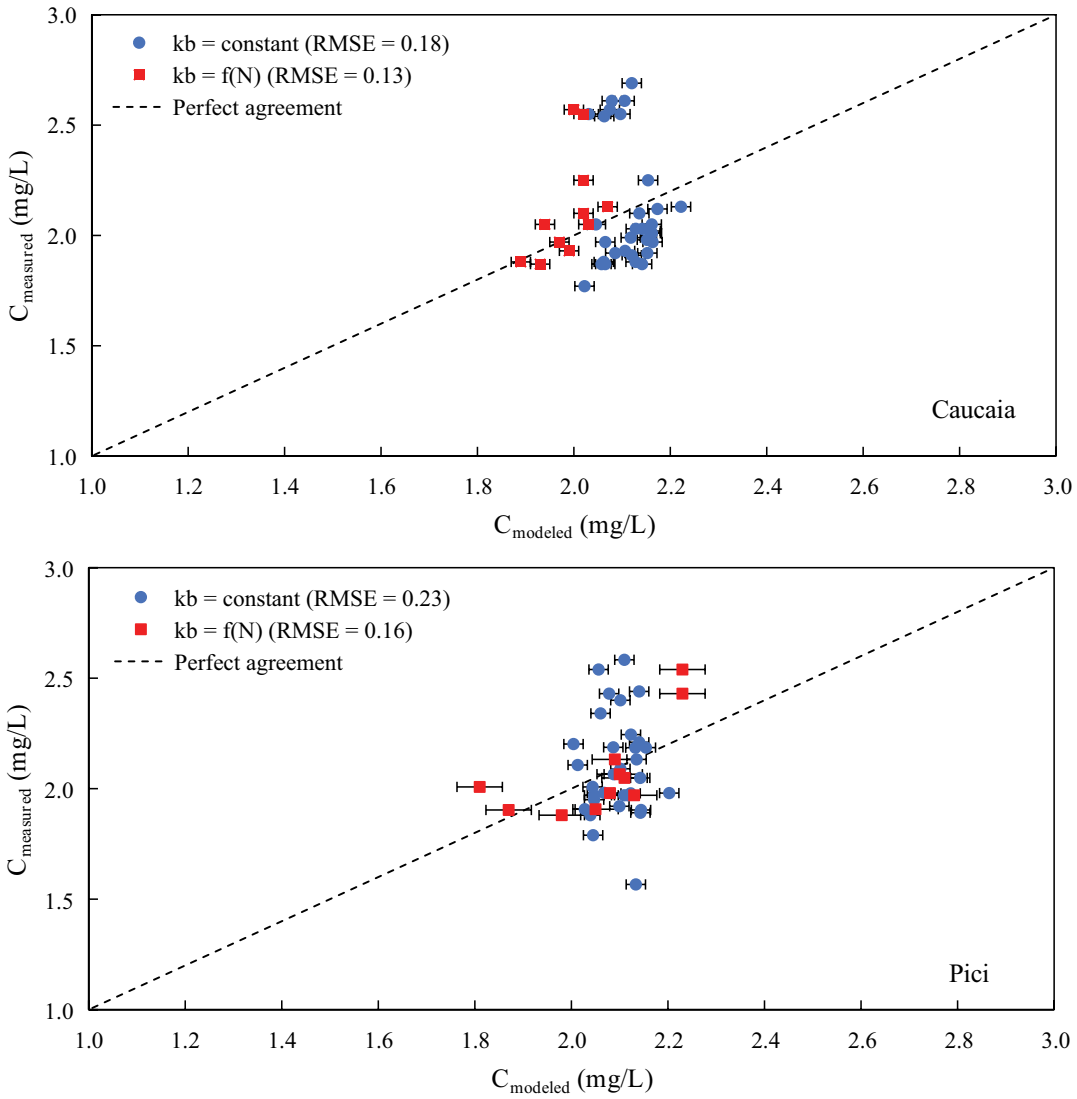

Fig. 4. Comparison between modeled and measured chlorine concentrations for both pipelines (Caucaia and Pici), considering two decay conditions: (1) a constant value of k_b = 0.2 d^{-1} for both pipelines and (2) a variable k_b as a function of nitrogen concentration (N): [k_b = 0.4824(N) – 0.4277]. 95% confidence intervals are represented by error bars.

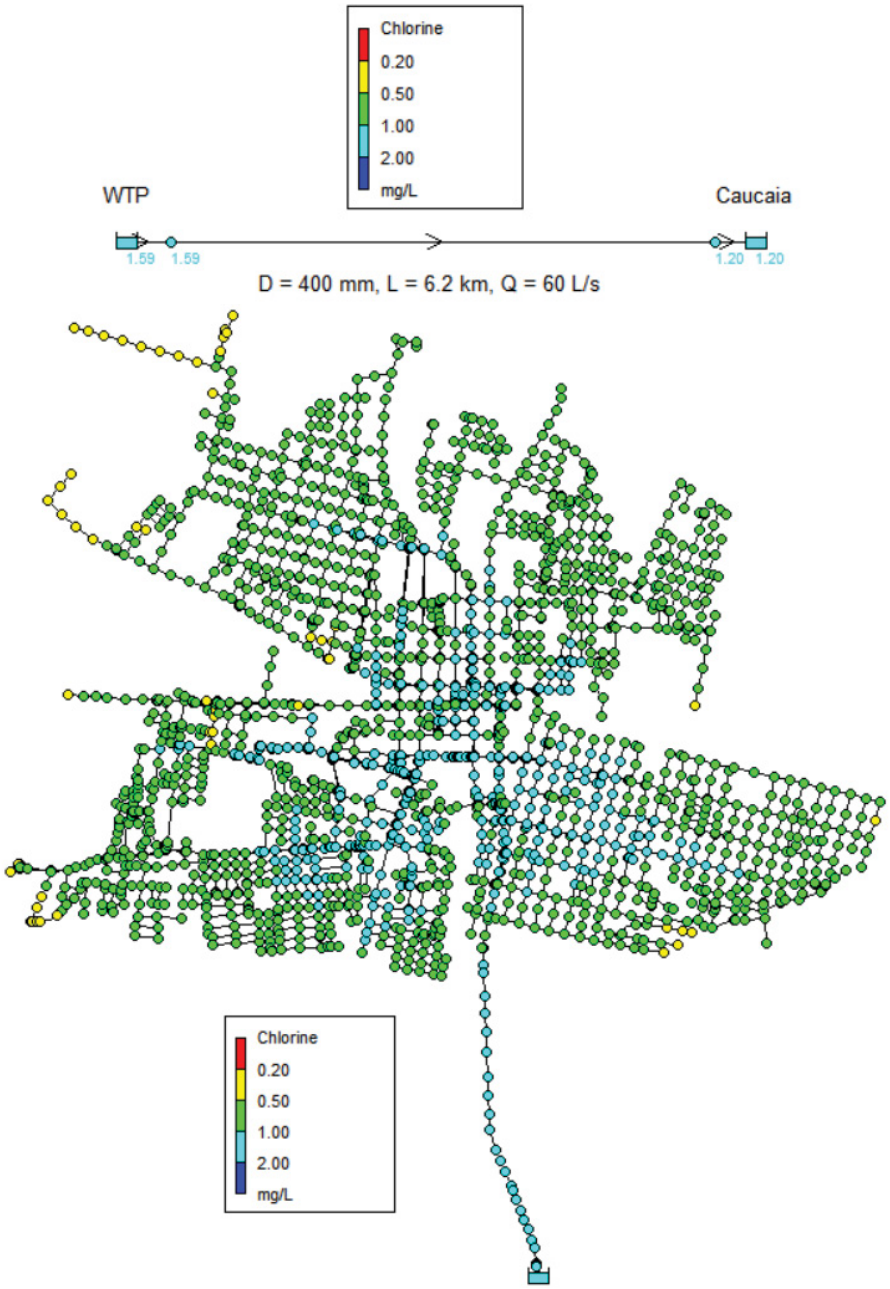

Fig. 5. Optimization of the operation of the water transmission (upper) and distribution (lower) systems of Caucaia for a scenario of low nitrogen concentration (N = 1 mg/L and k_b = 0.06 d^{-1}).

(N): [k_b = 0.4824(N) – 0.4277] (Fig. 3). Note that 95% confidence intervals are represented by error bars. The results indicate that, while the constant k_b condition yielded root-mean square errors (RMSE) of 0.18 and 0.23 mg/L for the pipelines of Caucaia and Pici, respectively, the variable k_b model provided RMSE of 0.13 and 0.16 mg/L, respectively. This indicates that the use of $k_b = f(N)$ improves model predictions. It is important to mention, however, that more detailed approaches such as the two-reactant (2R) or augmented two-reactant (2RA) decay models of Fisher et al. [12,13] have potential to better represent the experimental results. However, in addition to the lack of detailed data for applying the 2R/2RA models, the single-reactant model proposed herein yielded results which were accurate enough (deviations of up to about 7%) for water resources management purposes, so that this simpler model could be easily embedded in EPANET or other softwares and incorporated by water utility companies. Note that these companies normally assume constant values for the bulk decay coefficient, which disregards the variability of water quality at the source. On the other hand, the 2R/2RA models present systems of equations and multiple parameters

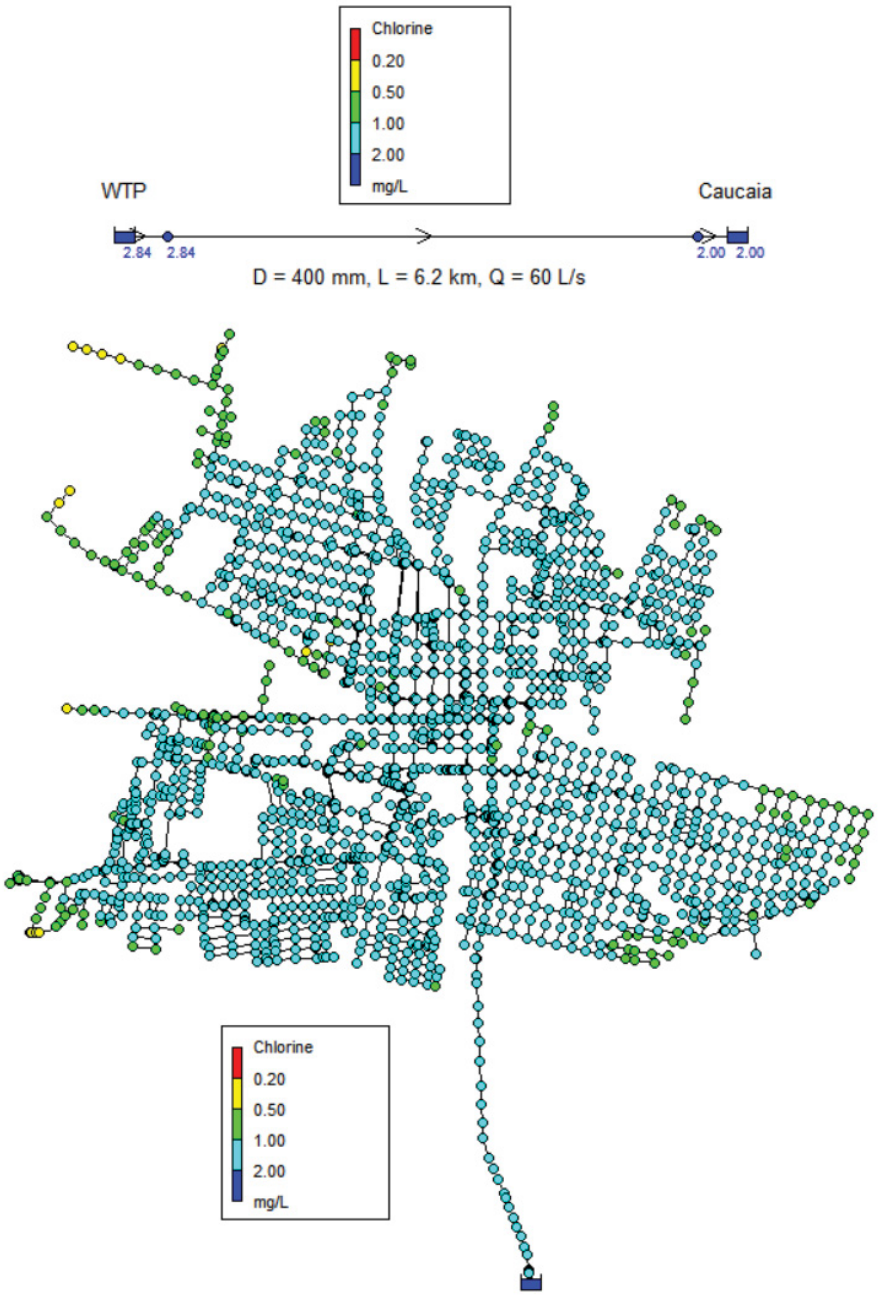

Fig. 6. Optimization of the operation of the water transmission (upper) and distribution (lower) systems of Caucaia for a scenario of high nitrogen concentration (N = 2 mg/L and k_b = 0.54 d^{-1}).

which make their calibration/solution procedures more complex than those of single-reactant models.

As an application of the results obtained in the present study, Figs. 5 and 6 show simulations of chlorine decay in the transmission pipeline as well as in the WDS of Caucaia by using the two correlations proposed here: $k_w = -0.5/\log(\varepsilon/D)$ and $k_b = 0.4824(N) - 0.4277$. Note that minimum chlorine concentrations from extended period simulations (48 h) are shown in the figures. While Fig. 5 considers as a first scenario the lowest observed nitrogen concentration (N = 1 mg/L and k_b = 0.06 d^{-1}), Fig. 6 considers a second scenario of high nitrogen concentration (N = 2 mg/L and k_b = 0.54 d^{-1}). The idea was to optimize the operation of both the transmission and distribution systems regarding chlorine dosage at the WTP as a function of N, without the need for rechlorination along these systems. In the first scenario (Fig. 5), it is clearly seen that C = 1.59 mg/L is enough to meet the management goal of 0.2 mg/L at the water distribution network extremities. On the other hand, in the second scenario (Fig. 6), C = 2.84 mg/L is necessary to reach this management goal. Hence, instead of using an approximately

constant chlorine concentration of C = 2.70 mg/L at the WTP outlet, as currently done by the Water and Sewage Company of Ceará (CAGECE) (Fig. 2), different values of C may be used (1.59–2.84 mg/L) to optimize the operation of the water supply system, depending on N concentration at the source waters.

4. Conclusions

This study analyzed the chlorine concentration upstream and downstream in two water mains of different diameters, materials and flow conditions in the Metropolitan Region of Fortaleza, Brazil, in order to investigate both experimentally and numerically the chlorine decay along these pipelines. Water quality at the source waters was also analyzed. Additionally, simulations of chlorine disinfection in a water distribution system were performed to illustrate the applicability of the results. The following conclusions can be drawn:

- The fitted values for the bulk decay constant (k_b = 0–0.8 d^{-1} for both water supply pipelines) and the wall decay constant (k_w = 0.2 and 0.1 m/d for the water supply pipelines of Caucaia and Pici, respectively) were within the ranges reported in the literature for similar flow conditions, pipe dimensions and materials.
- The time-variations of chlorine concentration (C) suggest that nitrogen (N) concentration in the source waters has a significant impact on chlorine decay. This has been attributed to additional chlorine demand due to potential reactions with nitrogenous material such as ammonia. On the other hand, as expected, chlorine decay did not correlate with phosphorus (P) concentration.
- Two correlations were proposed for prediction of the bulk decay constant [$k_b = 0.4824(N) - 0.4277$] and the wall decay constant [$k_w = -0.5/\log(\varepsilon/D)$].
- The use of the proposed correlation for k_b better fitted the experimental data than assuming a constant value (k_b = 0.2 d^{-1}).
- Instead of using an approximately constant chlorine concentration (C = 2.70 ± 0.06 mg/L) at the WTP outlet, as currently done by the water utility company, simulations revealed that the management goal of 0.2 mg/L can be achieved at network extremities, without the need for rechlorination along the system, by setting C to values ranging from 1.59 to 2.84 mg/L, depending on N concentration.
- The results obtained in the present study are important to optimize chlorine disinfection in typical water supply systems in Brazil and other tropical/developing countries, which present similar chlorine/nitrogen concentrations and hydraulic conditions.

Data availability statement

All data that support the findings of this study are available from the corresponding author upon reasonable request.

Acknowledgments

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