Comparison of commercial disinfectants and an in loco-produced solution: free residual chlorine decay in human supply waters

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ABSTRACT

Disinfection process is used in the treatment of water for human supply to promote sanitary safety and provide users with drinking water that meets potability standards. Thus, it is necessary to sustain a minimal concentration of free residual chlorine (FRC) throughout the entire distribution system. The present study investigated the decay process of FRC concentration in water destined for human supply. The decay was evaluated in bench-scale testing, using sodium hypochlorite, calcium hypochlorite, sodium dichloroisocyanurate (organic chlorine) as disinfectant agents, and also an alternative disinfectant solution (ADS) produced in loco, with oxidizing and disinfectant properties, which is being used in Brazilian sanitation industry. To evaluate the decay, four models were fitted: first-order, nth-order, limited first-order and parallel first-order, hence determining the corresponding parameters which describe the decay speed of the FRC concentration in water. Achieved results demonstrated that all models were statistically significant and predictive. However, parallel first-order model produced the best fit. Regarding the evaluated disinfectants, it was noted the preeminence of ADS solution when compared to the others, since it imparted a higher FRC over time, a behavior indicated by lower values for reaction rate constant in all models and when compared to other disinfectants used in this study.

Key words: chlorine decay, kinetic models, water disinfection

HIGHLIGHTS

- Alternative disinfectant solution (ADS) tends to maintain a greater residual chlorine over time.
- Chlorine decay over time is best described by the first-order parallel model.
- The use of disinfectants that contain not only chlorine derivatives guarantee a greater residual free chlorine over time.

INTRODUCTION

For water to play its role in society, it is necessary to take a lot of care, from the abstraction in springs and aquifers to the use and disposal by the population (Salvatierra et al. 2010). Such care is related not only to aspects of water quality, but also to safety regarding its availability to meet human, industrial, commercial, agricultural, livestock, recreational, electricity generation, among other supplies. Thus, the distribution must be carried out rationally, avoiding degradation and water losses (Santos et al. 2011).

In Brazil, the procedures for the control and surveillance of water quality and potability standards for human consumption were reviewed and established in Consolidation Ordinance No. 5, Annex XX, of September 28, 2017 of the Ministry of Health (Brasil 2017).

To protect public health, disinfection is applied to water with the secondary objective of maintaining a disinfectant residual throughout the distribution system, ensuring this residual even at the ends of the networks, limiting microbial growth (Silva et al. 2019).

The chlorination process introduced in the early nineteenth century aims to reduce the spread of diseases through water. It decisively contributed to this purpose; however, due to the use of chlorine as the main disinfectant in water supply systems around the world, microorganisms acquired resistance to it over time, enabling outbreaks of water-borne diseases (Souza &
When applied in the form of gas, chlorine poses high risk of leakage; because of that, it has been replaced by other less dangerous products.

A proven technology available in the Brazilian market is the in loco sodium hypochlorite generator, which uses water, salt (sodium chloride) and electricity as inputs. The production of this disinfectant agent includes the electrolysis of sodium chloride in brine prepared at a concentration of 3%, which is soon after subjected to a direct current in an electrolytic cell containing anode and cathode (Pacheco et al. 2018). The result of the electrolytic reaction of sodium chloride is a solution that contains hypochlorite ion, hypochlorous acid, other chlorine species and traces of other oxidizing agents such as hydrogen peroxide, hydroxyl radicals and oxygen free radicals. The solution can be produced continuously, dosed directly in water, or in batch (Garcia 2018).

When it comes into contact with water, chlorine undergoes hydrolysis and this reaction results in the hypochlorite ion and hypochlorous acid, substances that are responsible for the free residual chlorine (FRC) content in the waters for human supply. The control of water pH is an important factor for disinfection, since hypochlorous acid is a weak acid and the main disinfectant agent, showing bactericidal power up to 80 times greater than that of hypochlorite ion. Its predominance occurs at pH between 2 and 8 (Sanabria & Julio 2013; Libânio 2016).

Due to the diversity of water characteristics, FRC species react with various substances such as iron, manganese, ammonia (inorganic matter) and organic matter in general, which causes two stages of decay of their concentrations: first reacting with inorganic material and then with organic material (Vieira et al. 2004; Gallandat et al. 2019; Nono et al. 2019).

Over the years, several studies on FRC decay have been conducted seeking to effectively describe, through mathematical models, the behavior of decay in various water supply systems for human consumption and which model has the best fit in these systems, reflecting the studied conditions (Brown et al. 2011; Fisher et al. 2011).

The first-order model, due to its simplicity, has been widely used in the simulation of the FRC decay in water supply systems (Rossman et al. 1994). For Munavalli & Kumar (2004), FRC decay is defined and it may be appropriate to admit first-order reactions in the models. However, several authors reported that this kinetic approach is often not the most accurate to describe the decay of chlorine concentration for water used in public supply (Clark 1998; Kastl et al. 1999; Jonkergouw et al. 2009; Monteiro et al. 2014). Other models were proposed, besides first-order kinetics, aiming at a description with greater accuracy of the decay of chlorine concentration in water (Sanabria & Julio 2013).

According to Vieira et al. (2004), the first parallel order model is the one that best adjusted to the consumption of FRC in the liquid mass by presenting two stages of FRC degradation, the first being the fastest, where reactions with inorganic matter and the second slower stage occur, in which chlorine reacts with the organic material present in water.

According to Figueiredo (2014), the factors that influence chlorine decay, such as natural organic matter concentration, initial chlorine concentration, temperature and hydraulic conditions, need to be added to the models, in order to make them more robust.

In this perspective, the aim of this study was to evaluate the decay of FRC concentration over time and to determine the kinetic coefficients of FRC decay for four disinfectant agents: sodium hypochlorite, calcium hypochlorite, sodium dichloroisocyanurate (organic chlorine) and an alternative disinfectant solution (ADS) produced in loco, in view of the need for conducting studies that evaluate the efficiency of new disinfectant agents and disinfection processes alternative to chlorine, which contribute to guaranteeing the quality of water for supply and allow greater safety to the communities served.

**METHODS**

**Experimental conditions and procedures**

The water used in the experiments came from the Water Treatment Plant of Gravatá (WTP-Gravatá) and was collected shortly after the filtration stage. Located in the municipality of Queimadas, PB, Brazil, and responsible for the treatment of the waters of the Epitácio Pessoa reservoir (Figure 1), this station is of the conventional type and has the following configuration: Parshall trough, mechanical flocculants, horizontal flow decanters, gravity filters, chlorine contact tank and a complete chemical treatment facility, with the use of aluminum sulfate as coagulant agent and chlorine gas in the disinfection step.

As WTP-Gravatá operates in full cycle, upon arriving at WTP, the raw water receives hydrated lime for pH correction, then in the coagulation stage, the aluminum sulfate is added in the Parshall trough, where the rapid mixing occurs. Then the water
proceeds to the stages of flocculation, decanting, filtration and disinfection with gaseous chlorine. Finally drinking water is distributed.

Thus, this station is responsible for supplying nine municipalities in the state of Paraíba, including Campina Grande, which has a medium-sized supply system and benefits 156,298 economies, through 135,532 home connections (Nascimento et al. 2016) with an estimated population of 409,731 inhabitants for the year 2019 (IBGE 2020).

Figure 1 | Location of the Epitácio Pessoa Reservoir (a) and WTP-Gravatá (b).
Some physical-chemical and microbiological characteristics of the water used to conduct the tests are presented in Table 1, with their respective average values.

In the present study, the disinfectant agents used were sodium hypochlorite, calcium hypochlorite, sodium dichloroisocyanurate (organic chlorine) and the agent produced from an electrochemical reactor, which will be described later and which, for discussion purposes here, will be called ADS (Alternative Disinfectant Solution), in order to verify the decay of its FRC species in the water volume.

The experiments were carried out as follows: (i) 5-L samples of filtered water were added in gallons with the same volume; (ii) doses of the different disinfectant solutions were added until reaching FRC concentrations of approximately 5.0 mgCl₂.L⁻¹, simulating the value maximum value allowed by Consolidation Ordinance No. 5, in Annex 7 of the Annex XX, of the Ministry of Health, at the exit of the water treatment station; (iii) FRC was determined at regular time intervals. The experiments were conducted at room temperature. Figure 2 illustrates the experimental apparatus of the decay test.

As illustrated in Figure 2, the experiments were conducted in triplicate for each disinfectant agent. Each test had a total duration of 1,440 minutes, with FRC determinations performed at times of 0, 10, 20, 40, 80, 160, 720 and 1,440 minutes. In parallel, the determinations of FRC were performed the monitoring of pH and temperature, in order to verify the variability of these parameters during the execution of the experiments, since these parameters are directly related to the degradation of FRC in water (Gallandat et al. 2019; Li et al. 2019; Goodarzi et al. 2020).

FRC concentrations were determined using the DPD-SFA titrimetric method, as described in the Standard Methods for the Examination of Water and Wastewater (APHA et al. 2012). Temperature and pH monitoring during the experiments was performed with a MS TECNOPON® mPA-210 benchtop pH meter.

**Alternative disinfectant solution (ADS) produced in loco**

The solution with disinfectant potential is produced from the electrolysis process in aqueous medium of sodium chloride (non-iodized common salt).

| Parameter         | Value              | Method                        |
|-------------------|--------------------|-------------------------------|
| Turbidity         | 0.71 NTU           | Nephelometric                 |
| Aparente color    | 12.30 uH           | Colorimetric                   |
| True color        | 8.90 uH            | Colorimetric                   |
| Absorbance 254 nm | 0.135              | Ultraviolet Absorption Method  |
| pH                | 7.51               | Potentiometric                 |
| Alkalinity        | 23.0 mgCaCO₃.L⁻¹   | Titrimetric                    |
| Total hardness    | 99.0 mgCaCO₃.L⁻¹   | Titrimetric                    |
| Total coliforms   | Presence           | Chromogenic substrate          |
| *E. Coli*         | Absence            | Chromogenic substrate          |

**Figure 2** | Experimental apparatus.
The reactor produces *in loco* a solution that contains sodium hypochlorite and hydrogen peroxide, with oxidizing potential greater than that of commercial sodium hypochlorite. Figure 3 illustrates in a schematic way the production process and its respective chemical species.

In this study, the static chlorine generator Hidrogeron® GE-15 was used. The generator’s operation to produce the solution (Figure 4) is started with the addition of 15 liters of water (Figure 4(I)), preferably slowed down to avoid electrodeposition of calcium and magnesium salts in the electrodes, with subsequent addition of 600 g of non-iodized sodium chloride (salt) (Figure 4(II)) and homogenization of brine solution (Figure 4(III)). Then, the reactor is closed and connected to the electrical power source. After 8 hours, the source is turned off automatically and the ADS is ready (Figure 4(IV)). At the end, the valve

![Figure 3](image1.png)  
**Figure 3** | ADS production process via electrolysis. *Source: Dantas et al. (2017).*

![Figure 4](image2.png)  
**Figure 4** | Operation of static chlorine generator for ADS production.
(A) that connects the upper and lower chambers is opened so that the solution is collected by the output valve (B) of the lower reservoir (Figure 4(V)). With this, the upper reservoir is emptied and a new batch can be produced.

**Determination of the kinetic coefficients of FRC decay and data analysis**

The kinetic coefficients of FRC decay were determined by fitting first-order, nth-order, limited first-order, parallel first-order models (Equations of 1, 2, 3 and 4, respectively) to the experimental data, based on the least squares method, and the parameters of each model were estimated by minimizing the sum of the residuals. These models were proposed by Haas & Karra (1984) to describe the decay of chlorine in the disinfection of wastewater, but have also been reported in the literature as capable of representing FRC decay in supply waters.

\[
C = C_0 e^{-kt} \quad \text{1st order} \\
C = \left( \frac{kt(n-1)}{C_0} \right)^{1/n} \quad \text{nth order} \\
C = C^* + (C + C^*)e^{(-kt)} \quad \text{1st limited order} \\
C = xC_0e^{(-k_1t)} + (1-x)C_0e^{(-k_2t)} \quad \text{1st parallel order}
\]

where:

- \( C \) = FRC concentration at a given time, in mg·L\(^{-1}\);
- \( t \) = time, in min;
- \( k \) = chlorine decay rate constant, in min\(^{-1}\) for the first-order and limited first-order models and in mg\(^{1-n}\)·L\(^{n-1}\)·min\(^{-1}\), for the nth-order model;
- \( C^* \) = chlorine limit concentration, in mg·L\(^{-1}\);
- \( n \) = reaction order, dimensionless;
- \( C_0 \) = initial concentration of FRC, in mg·L\(^{-1}\);
- \( x \) = FRC fraction, dimensionless;
- \( k_1 \) and \( k_2 \) = respectively, fast and slow chlorine decay rate constants, in min\(^{-1}\).

The adequacy of the models to the decay data was evaluated through the coefficient of determination (R\(^2\)) and by the Fisher test (F test). For the fit of the models to the observed data, the statistical analyses were performed using Statistica software version 12.0 (Statsoft 2011).

**RESULTS AND DISCUSSION**

The decay behavior of the FRC concentration over time, for each disinfectant, was used to determine the kinetic coefficients of FRC decay in the volume of water, which describe the speed at which chlorine reactions occur in water destined for human supply.

Hypochlorous acid and hypochlorite ion are the main oxidizing agents that make up the FRC, and react with other substances present in the water, thus, once added to water, chlorine initiates several interactions resulting in the decay of its concentration. Several substances present in water is able to react with chlorine simultaneously at different speeds. Inorganic substances such as ammonia, nitrite, iodides and bromides, sulfites, iron (II) are responsible for interacting with chlorine at faster speeds, while manganese (II) reacts more slowly. In turn, because they present more complexity and variety of species, organic substances present reaction velocities with chlorine from fast to very slow, according to their composition (Deborde & Vonguten 2008; Sanabria & Julio 2013).

Figure 5 illustrates the FRC decay curve for the different disinfectant agents evaluated. It is observed that the free chlorine residual in the water mass tends to be consumed more quickly in the first minutes of the test, reaching a greater stability of its concentration over time. This is due to water quality conditions, as organic and inorganic substances will react with FRC and generate higher initial consumption. After all these substances are oxidized, the FRC will tend to decay more slowly. This behavior is consistent with what is observed in other studies available in the literature (Gallandat et al. 2019; Nono et al. 2019).
Among the decay curves illustrated in Figure 5 for the different disinfectant agents, it is noticed that the ADS shows a more stable behavior, with a lower decay of the portion of rapid consumption at the beginning and a higher concentration of FRC throughout the test.

The water used in the tests has an apparent color of 12.30 uH and true color of 8.90 uH, however a low absorbance value was verified 254 nm (Table 1) that indicates low concentration of organic matter in suspension. According to the obtained values, the true color represents about 70% of the apparent color, indicating that dissolved solids and colloidal particles in water are in greater proportion than suspended solids, and this apparent color value may represent the by-products of the decomposition of organic matter in the reservoir, as well as the possible presence of dissolved inorganic substances, such as iron, manganese and chemicals, such as lime hydrates and aluminum sulfate, used for pH correction and coagulation, in the process of water treatment (Libânio 2016).

Wang et al. (2020) comment that the presence of biofilms is frequent in water supply systems and results mainly from organic matter not removed in conventional water treatment and from the aging of the distribution network. The authors point out that the control of the biofilm depends heavily on the residual levels of disinfectant in the system and that FRC values equal to or below 0.5 mg·L⁻¹ are not effective for this purpose. Based on this, the present study shows the potential of ADS for greater network security, since, even after 1,440 min, the FRC level in water was above 1.0 mg·L⁻¹.

In relation to the parameters pH and temperature, Figure 6 illustrates their variability for the data obtained in the decay tests with the disinfectant agents evaluated. Based on these parameters, and on the FRC concentration present in water, the Consolidation Ordinance No. 5, Annex XX of September 28, 2017 of the Ministry of Health, establishes the minimum contact time for disinfection performed through chlorination.

Water pH is a factor that significantly influences the efficiency of disinfection with chlorine compounds, as it promotes the dissociation of hypochlorous acid (HOCl) and the formation of hypochlorite ion (OCl⁻). The sum of the HOCl and OCl⁻ species constitutes the FRC, with OCl⁻ being the weakest disinfectant and prevailing at higher pH (>8.0), while HOCl is more reactive and prevails at lower pH (<8.0) (Libânio 2016).

For this reason, research studies indicate that pH is a parameter that strongly influences the rate of chlorine decay, as well as the stability of the disinfectant and the formation of disinfection by-products (Gallandat et al. 2019; Li et al. 2019). In the present study, the decay of FRC for the four disinfectant agents occurred at pH values lower than 8.0, without large variations, which according to Libânio (2016) ensures a prevalence higher than 80% of hypochlorous acid.

Chlorine decay is a reaction that also depends directly on temperature. Thus, the higher this parameter, the faster the FRC self-decay will occur (Li et al. 2019; Goodarzi et al. 2020). This observation was confirmed by Kim et al. (2019), who found...
that the decay of FRC in supply water occurred more accelerated at 23 °C than at 18 °C. However, in the present study, no significant variations in temperature were observed between the tests.

An alternative disinfectant that has been studied is peracetic acid, considered quite efficient for water disinfection. Its behavior in the supply water was evaluated by Zhang et al. (2020), who observed decay of 10% from an initial concentration of 5.0 mg·L⁻¹ after 60 min in water with pH of 7.0–7.5 and temperature of 20 °C. Under similar conditions of initial concentration, time and pH, in this study, the decay of ADS was similar to that of peracetic acid, even with the higher water temperature (24 °C), which highlights its stability.

First-order, nth-order, limited first-order and parallel first-order models were fitted to the data obtained from the FRC decay experiments in the water volume.

The first-order model (Equation (1)) is widely applied in studies on chlorine decay in water distribution systems (Maier et al. 2000; Sanabria & Julio 2013; Rodrigues & Scalize 2019), where it is assumed that the concentration, $C$, of FRC decreases exponentially over time, $t$, and $k$ is the decay rate constant of chlorine.

For the limited first-order model (Equation (3)), $C^*$ is considered a part of the initial concentration that does not react with no species of organic or inorganic nature present in the water, and the first-order exponential decay is dependent on the complementary part ($C_0-C^*$). In the nth-order model (Equation (2)), the decay rate constant is proportional to the umpteenth power of the concentration $C$.

In turn, the parallel first-order model (Equation (4)) assumes that there are two specific rate constants: one part ($xC_0$) of the initial chlorine concentration decays exponentially with specific rate constant $k_1$, and the other ($1-x)C_0$ decays exponentially with specific rate constant $k_2$.

Figure 7 presents the fits of the models to the experimental data, where it is possible to verify that the parallel first-order model showed the best fit for all disinfectant agents.

Some authors use the correlation coefficient ($R$) as a criterion for hierarchization in the process of choosing the best kinetic model (Powell et al. 2000; Vieira et al. 2004; Beleza 2005). However, here, to evaluate the best fit, a statistical analysis was performed, adopting as a criterion for choosing the best model the highest values of the coefficients of determination ($R^2$) and $F$ test.

Table 2 shows the statistical values of $R^2$ and $F$ test, as well as the values of the parameters fitted for each model analyzed. Although all models showed satisfactory fits, with coefficients of determination above 0.83 and $F_{\text{calculated}}$ higher than $F_{\text{tabulated}}$, the parallel first order model presented the best values for the $F$ test ($F_{\text{calculated}} > F_{\text{tabulated}}$) and higher values of the coefficients of determination, when compared the models for each disinfectant agent. The model is statistically significant if $F_{\text{calculated}} > F_{\text{tabulated}}$ and, if the $F_{\text{calculated}}/F_{\text{tabulated}}$ ratio ($F$ test) is greater than 10, the model is not only statistically significant but also predictive (Barros Neto et al. 2001), which can be observed for all models evaluated for different experiments with 95% confidence interval.
An accurate analysis of the parallel first-order model, in relation to the adjustable parameters $k_1$ and $k_2$, reveals the coherence of the results obtained ($k_1 > k_2$), since these coefficients describe the speed of fast and slow reactions of FRC in water, respectively. The high values of $k_1$ are justified by the direct relationship of this parameter with the reactions between chlorine and inorganic substances present in water, which are more easily degraded than organic ones.
Thus, the high values of $k_1$ obtained in the fit of the models to the data are coherent. In addition, the value of the kinetic constant associated with reactions involving organic compounds in the raw water of the water supply system of Campina Grande is 5% (Nascimento et al. 2016), attesting that inorganic compounds are responsible for most of the chlorine demand.

Table 3 presents the analysis of variance for the parallel first-order model, which showed the best values of $R^2$ (>0.99) and $F$ test (152.9764). It is observed that the disinfectant agent ADS was superior to the others, mainly in relation to the components of the $F$ test.

Regarding the FRC decay rate in the water volume, for the parallel first-order model, it can be observed that the values of the kinetic constants of reaction speed ($k_1$ and $k_2$) were lower for ADS. That is, for this disinfectant agent the concentration of FRC decays more slowly, leaving a greater residual over time and, consequently, making the water safer for consumption by users of the supply system.

This fact can be attributed to the characteristics of the chemical species that make up the ADS solution: hydrogen peroxide, which has a great oxidative power capable of oxidizing substances present in water, and sodium hypochlorite, which is responsible for maintaining FRC for longer in the water mass.

By presenting a higher oxidation power than the oxidizing agents present in THE CRL, hydrogen peroxide will oxidize the organic and inorganic substances present in water. Normally these substances would be oxidized by the chlorine plots, resulting in a higher consumption, and consequently, a greater decay of their concentration in water. Due to the presence of hydrogen peroxide in the SDA, the CRL concentration remains higher throughout the distribution system, making it possible to reach points farther from the network, avoiding the need for a new disinfectant dosage.

Thus, lower doses of disinfectant can be applied in the disinfection stage of the treatment, keeping the network within the safety standards established by Consolidation Ordinance No. 05/2017, Annex XX, which establishes that at any point in the public network there should be at least 0.2 mg·L$^{-1}$ of FRC. This reduction in dose can reduce the operating costs. A cost-benefit economic analysis was performed by Pacheco et al. (2018), in order to verify the financial viability in the replacement of chlorine gas by sodium hypochlorite produced in loco in a water treatment plant in the city of Uberlândia, Minas Gerais, and these authors found an average reduction of 32.22% in the values paid monthly with disinfectant. However, it is necessary to conduct future studies to assess the cost reduction related to the variation of the dose applied to maintain the FRC throughout the supply network.

**CONCLUSIONS**

The results presented in this article suggest that the adjustable parameters of the different models evaluated indeed represent the decay of FRC in human water supply, since all were statistically significant and predictive. Therefore, they can be used in future studies of modeling of the disinfection process applied to the treatment of water for human consumption.

The parallel first-order kinetic model proved to be the most appropriate model to describe the decay of FRC for the different disinfectant agents tested in this study.

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**Table 3 | Analysis of variance for the parallel first-order model applied to different disinfectant agents**

| Desinfectant Source of variation | SS       | DF | MS        | $F$ calculated | $F$ tabulated | Test F | $R^2$ |
|---------------------------------|----------|----|-----------|----------------|---------------|--------|-------|
| ADS Regression                  | 127.8218 | 3  | 42.6073   | 8,318.4800     | 5.4095        | 1537.7678 | 0.9983 |
| Residual                        | 0.0256   | 5  | 0.0051    |                |               |        |       |
| Total                           | 127.8474 | 7  |           |                |               |        |       |
| Sodium hypochlorite Regression  | 72.3643  | 3  | 24.1214   | 827.5181       | 5.4095        | 152.9764 | 0.9901 |
| Residual                        | 0.1457   | 5  | 0.0291    |                |               |        |       |
| Total                           | 72.5100  | 7  |           |                |               |        |       |
| Calcium hypochlorite Regression | 92.3991  | 3  | 30.7997   | 1,005.8717     | 5.4095        | 185.9471 | 0.9925 |
| Residual                        | 0.1531   | 5  | 0.0306    |                |               |        |       |
| Total                           | 92.5522  | 7  |           |                |               |        |       |
| Sodium dichloroisocyanurate     | 56.7252  | 3  | 189.084   | 2,098.7451     | 5.4095        | 387.9774 | 0.9975 |
| Residual                        | 0.0450   | 5  | 0.0090    |                |               |        |       |
| Total                           | 56.7702  | 7  |           |                |               |        |       |

Thus, the high values of $k_1$ obtained in the fit of the models to the data are coherent. In addition, the value of the kinetic constant associated with reactions involving organic compounds in the raw water of the water supply system of Campina Grande is 5% (Nascimento et al. 2016), attesting that inorganic compounds are responsible for most of the chlorine demand.
The ADS solution presented itself as a disinfectant agent capable of maintaining a free chlorine residual in water for a long period of time and at a small dose, ensuring the residual within the established by the Ministry of Health for potability, in addition to promoting greater sanitary safety of the water supplied to users of the supply system.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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