Chlorine Decay Kinetics in Surface Waters of Angola

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Abstract

Chlorine is frequently chosen as the primary disinfectant for producing potable water. During chlorination, chlorine reacts with organic matter present in water and is consumed over time. Knowing how fast this decay occurs is important for designing contact tanks and selecting the right chlorine dose. This study investigated chlorine decay in raw water from the Bengo River, Angola, collected at the intake of the Kifangondo Water Treatment Plant. Experiments were carried out at 25 °C using the bottle test method. The initial chlorine concentration was varied between 0.65 and 4.7 mg Cl_2 L^{-1}, while the organic matter concentration was kept constant at an oxidability of 4.6 mg O_2 L^{-1} (pH 7.5–7.6). Residual chlorine was measured at fixed time intervals over 20 minutes. Four kinetic models were tested: first-order (P1), pseudo second-order (P2), limited first-order (PL), and parallel first-order (PP). Non-linear optimization was used to fit the four models to the experimental data. The quality of each fit was evaluated using the coefficient of determination (R^2), the estimated variance (S^2), and the F-test at a 95% confidence level. Chlorine decay was fast in the first few minutes and slowed down afterwards, stabilising at approximately 20 minutes. The fraction consumed within this period ranged from 60% for the highest initial concentration to 97% for the lowest. The first-order model gave a poor fit for higher concentrations (R^2 = 0.72–0.74). The pseudo second-order model performed better at lower doses. The limited first-order model gave a good fit across all conditions (R^2 = 0.975–0.997). The parallel first-order model gave the best overall fit (R^2 = 0.985–0.999) and was statistically superior to the other models at C_0 = 4.7 mg Cl_2 L^{-1}. A practical example of contact tank sizing was also presented using the pseudo second-order model for the Kifangondo Water Treatment Plant.

Keywords: *Bengo River, chlorine, disinfection, kinetic modelling, organic load, surface water.*

1. Introduction

As a vital resource for all living organisms, water is central to humans. It is indispensable for urban and industrial needs, irrigation, and hydroelectricity, as well as for supporting biodiversity and leisure activities [1, 2].

With the improvement in the quality of life, there has been increasing focus on essential resources. In the past, people thought water would never end, but now we know it must be protected. This shift has made water quality a more important topic. In recent decades, rapid industrial growth and agricultural activity have altered the sources and quality of surface water. Agricultural expansion near rivers and lakes has increased the risk of pesticide contamination. Agriculture is one of the main causes of environmental pollution, being responsible for adding many types of pollution to the water [3].

Many cities in Angola get their water from different rivers and springs. In the capital, water comes mainly from two rivers. The Kifangondo plant, located in Cacuaco, treats water from the Bengo River. In the Viana area, the Kikuxi plant treats water from the Kwanza River.

Water quality is essential to make water safe for human use. Disinfection is needed to remove bacteria and prevent health risks. Chlorination is the most common method usually applied at the final stage of treatment. During this process, chlorine reacts with organic matter and is consumed. It also continues to react and decay in the distribution system.

At treatment plants, chlorine or sodium hypochlorite is added to the water at the start (pre-oxidation). This helps the next steps to perform better, especially the coagulation and flocculation process [4]. This pre-oxidation step makes the treatment process more effective. It helps remove contaminants and improves water quality for drinking.

Natural organic substances, mainly humic and fulvic acids, react with chlorine in the water [5]. Humic acids react faster than fulvic acids, so their levels drop quickly during chlorination. This reaction creates by-products, and the most common ones are trihalomethanes, like chloroform, dichlorobromomethane, chlorodibromomethane, and bromoform. Trihalomethanes pose potential health risks; therefore, quantification and control measures are required to ensure water safety [6, 7].

To design tanks for the pre-chlorination step and calculate the chlorine dosage, we must understand the chlorine use rate during initial oxidation in surface waters, as stated by Pérez et al. [8]. The rate of chlorine consumption depends on the water composition, the initial chlorine level, temperature, and other conditions. Laboratory tests help identify which decay model best describes how chlorine concentration changes over time [9]. The selected model and known kinetic parameters can then be used to size a pre-chlorination unit.

Water submitted to chemical coagulation and flocculation has very little organic matter. This makes it hard to study how fast chlorine is used. Even so, chlorine is often the first step to treat raw surface water.

This work focused on the decay kinetics of chlorine in water samples from Bengo River (Angola), at the Kifangondo water treatment plant's intake point, for the production of drinking water. The specific objectives were (1) to test some kinetic models on chlorine decay data, and (2) to compare and select the most suitable model.

2. Materials and Methods

2.1 Samples

The Bengo River near the Kifangondo Water Treatment Plant in Luanda, Angola, was the source of the surface water for this study (Fig 1). The samples were stored in 5 L plastic containers and sent to the Laboratory of Separation Engineering, Chemical Reaction, and Environment at Agostinho Neto University (LESRA) for testing. They were kept in a refrigerator at 4 °C until the tests were done.

Fig. 1. Bengo River, Kifangondo water treatment plant, Angola.

2.2 Chemicals and analysis methods

Commercial bleach containing 5.25% NaOCl was diluted in water to prepare the sodium hypochlorite stock solution. The final concentration was set at 64.3 mg Cl_2 L^{-1}.

The DPD titration method with ammonium ferrous sulfate was used to determine the residual chlorine, as described in Standard Methods for the Examination of Water and Wastewater [10].

A volumetric oxidation and reduction method was used to quantify organic matter in the raw water. The oxidability with potassium permanganate was measured to determine the oxygen tied to the organic matter based on the chemical consumption.

The Standard Methods guide was used to prepare the solutions of ammonium ferrous sulfate, phosphate buffer, and DPD indicator [10].

2.3 Chlorine decay modeling

In recent years, considerable research has focused on chlorine consumption in both surface water and drinking water, resulting in the development of various models to characterize chlorine decay [11-15]. The modeling of chlorine decay aims to understand the changes in chlorine concentration over space and time, accounting for its physical movement and the chemical reactions it undergoes with other species present in the solution.

In this study, the following models were chosen to investigate chlorine decay (Equations 1-4):

a) First order model (P1)
The rate of concentration decrease is proportional to the chlorine concentration remaining in solution.

$$C = C_0.e^{-kt} \tag{1}$$

b) Pseudo second-order model (P2)
The decay rate depends on the amount of chlorine remaining and the substances that react with chlorine.

$$C = C_0/(1 + C_0.K.t) \tag{2}$$

c) Limited first-order model (PL)
The decay of chlorine follows first-order kinetics until it reaches a limiting concentration.

$$C = C^* + (C_0 - C^*).e^{-kt} \tag{3}$$

d) Parallel first-order model (PP)
There are two first-order decay kinetics, one simulating the faster phase of decay and the other the slower phase.

$$C = C_0.x.e^{-k_1 t} + C_0.(1 - x).e^{-k_2 t} \tag{4}$$

where:
C is the chlorine level at time t, and C_0 is the starting level, both in mg L^{-1}
C^* represents the unreacted chlorine fraction in mg L^{-1}
k, k_1 and k_2 are the velocity constants per minute
K is the decay constant in L per mg per minute
x; fraction of initial chlorine

Chlorine decay modelling is important for designing chlorination systems. It helps determine the correct size of the contact tank so that water remains long enough for effective disinfection. It also helps select the appropriate initial chlorine dose to maintain a safe residual level. In addition, it ensures that CT requirements (Concentration × Time) are met according to water quality standards. Overall, this approach supports safer and more efficient system design.

Chlorine decay modelling helps operators manage chlorination units more effectively. It allows them to adjust the chlorine dose when water quality changes. This helps to avoid excessive use of chlorine and reduces costs. It also limits the formation of harmful by-products, such as trihalomethanes.

The model can also account for seasonal changes, rainfall or temperature variations. Most importantly, this ensures that safe chlorine levels reach the consumer taps.

2.4 Experimental procedure

The bottle test was chosen to study chlorine decay in the water. This method looks at reactions that happen only in the liquid. Water samples with a known starting chlorine concentration were put into Winkler bottles. The chlorine was measured at fixed times to check the reaction kinetics.

All tests were run in duplicate at 25 °C across two experiments, where the initial chlorine level was varied with constant organic matter in the first test, and the organic matter was varied with constant initial chlorine in the second test. Figure 2 illustrates the bottle test adapted from [16].

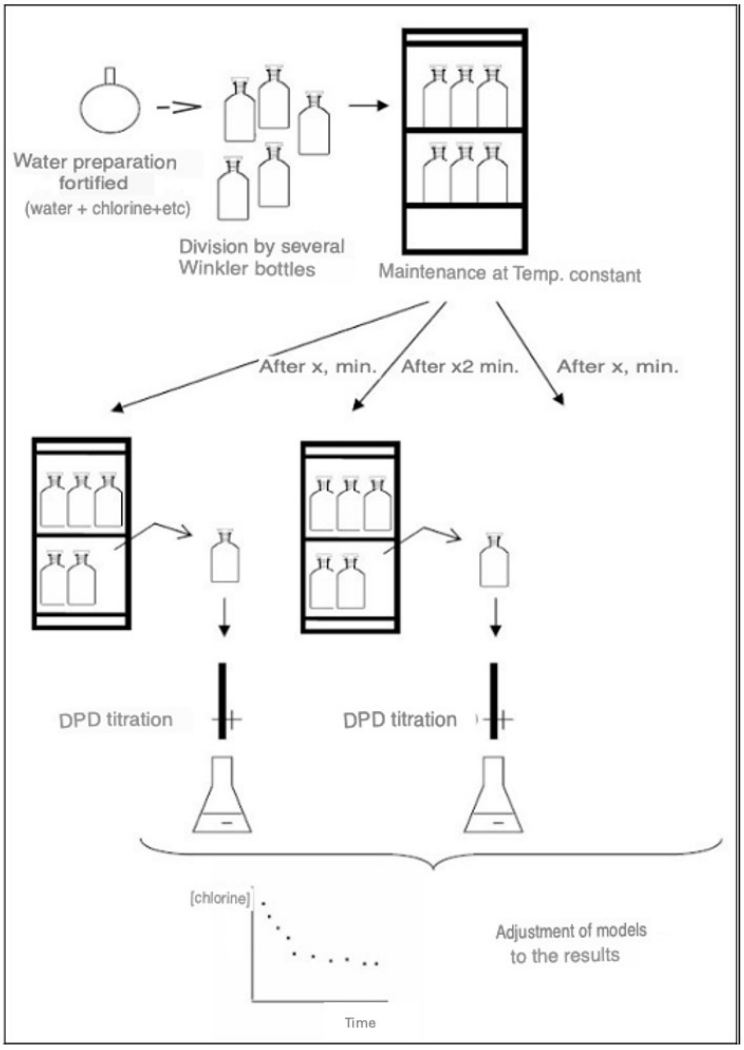

Fig. 2. Procedure for determining the kinetic decay constant into water (adaptaded from [16]).

3. Results and Discussion

3.1 Chlorine concentration 0.65 – 4.7 mg Cl_2 L^{-1} (T = 25 ºC)

Four chlorine decay experiments were carried out at 25°C using water from the Bengo River. The initial chlorine concentration was varied between 0.65 and 4.7 mg Cl_2 L^{-1}, while the initial organic matter concentration was held constant, with an oxidability of 4.6 mg O_2 L^{-1}. This oxidability indicates a moderate to high organic matter content, typical of surface water in tropical rivers such as the Bengo River in Angola, where natural organic load is often significant. In Portugal, a similar value would be considered normal for southern rivers, though slightly elevated for reservoirs in the centre and north of the country. Residual chlorine concentrations were measured at fixed time intervals and are reported in Table 1.

Tab. 1. Evolution of residual chlorine concentration over time at initial chlorine doses ranging from 0.65 to 4.7 mg Cl_2 L^{-1} (pH 7.5–7.6, oxidability 4.6 mg O_2 L^{-1}, T = 25 °C).

Time (min)	**C (mg Cl_2 L^{-1})**			
	C_0 = 0.65	C_0 = 1.3	C_0 = 2.7	C_0 = 4.7
0	0.65	1.3	2.7	4.7
3	0.20	0.60	1.55	2.9
6	0.15	0.40	1.50	2.4
9	0.05	0.30	1.25	2.2
12	0.02	0.25	1.20	2.1
15	0.02	0.20	1.20	2.05
20	0.02	0.15	1.10	1.95

The purpose of these experiments was to determine the kinetic rate constants for the decay of chlorine in water, thereby accurately predicting the rate at which reactions occur between chlorine and the organic matter present in raw water.

The experimental duration of 20 minutes proved adequate for all tests. By the end of this time, the free residual chlorine concentration had stabilized at a constant value, indicating that the reaction had reached a slow decay phase. The percentage of chlorine consumed within 20 min depended on the initial concentration, ranging from approximately 60% for the highest starting concentration (4.7 mg Cl_2 L^{-1}) to approximately 97% for the lowest (0.65 mg Cl_2 L^{-1}).

Four kinetic models were evaluated: first-order, pseudo second-order, limited first-order, and parallel first-order. The models were fitted to the experimental data using KaleidaGraph (version 4.1.3, Synergy Software), which applies the Levenberg-Marquardt algorithm for non-linear optimization. The estimated parameters for each model, as well as the coefficient of determination (R^2) and the estimated variance (S^2), are presented in Tables 2 to 5.

Tab. 2. Parameters of the first order model.

C_0 (mg Cl_2 L^{-1})	k ± SE	R^2	S^2
0.65	0.318 ± 0.036	0.980	$2.72x10^{-2}$
1.3	0.178 ± 0.020	0.933	$1.29x10^{-1}$
2.7	0.051 ± 0.014	0.725	$3.47x10^{-1}$
4.7	0.052 ± 0.014	0.741	1.08

Tab. 3. Parameters of the pseudo second-order model.

C_0 (mg Cl_2 L^{-1})	K ± SE	R^2	S^2
0.65	1.284 ± 0.199	0.986	$2.15x10^{-2}$
1.3	0.289 ± 0.004	0.999	$8.99x10^{-2}$
2.7	0.0429 ± 0.0069	0.874	$3.07x10^{-1}$
4.7	0.0248 ± 0.004	0.895	$9.32x10^{-1}$

Tab. 4. Parameters of the limited first-order model.

C_0 (mg Cl_2 L^{-1})	C^* ± SE	K ± SE	R^2	S^2
0.65	0.025 ± 0.017	0.365 ± 0.049	0.986	$2.87x10^{-2}$
1.3	0.194 ± 0.058	0.302 ± 0.026	0.994	$9.52x10^{-2}$
2.7	1.191 ± 0.138	0.393 ± 0.073	0.975	$1.64\ 10^{-1}$
4.7	2.035 ± 0.086	0.354 ± 0.021	0.997	$5.28x10^{-1}$

Tab. 5. Parameters of the parallel first-order model.

C_0 (mg Cl_2 L^{-1})	x ± SE	k_1 ± SE	k_2 ± SE	R^2	S^2
0.65	0.475 ± 0.356	0.999 ± 1.787 [a]	0.182 ± 0.080	0.993	$2.30x10^{-2}$
1.3	0.385 ± 0.136	0.999 ± 1.544 [a]	0.077 ± 0.021	0.985	$5.77x10^{-2}$
2.7	0.415 ± 0.041	0.926 ± 0.506	0.019 ± 0.006	0.990	$6.68x10^{-2}$
4.7	0.499 ± 0.005	0.444 ± 0.010	0.009 ± 0.001	0.999	$1.01x10^{-2}$

[a] no statistically significant

The first order model (P1) provided a reasonable fit for the two lowest initial concentrations (C_0 = 0.65 and 1.3 mg L^{-1}), with R^2 values of 0.980 and 0.933 (Table 2). For the higher concentrations, the fit was poor, with R^2 dropping to 0.70. This was expected, as the data clearly show a residual chlorine plateau that a simple first-order model cannot capture. In different studies had been reported that when initial chlorine varies or additional reactants are present [17, 18].

According to the data in Table 3, the pseudo second-order model performed well for C_0 = 0.65 and 1.3 mg L^{-1}, with R^2 of 0.986 and 0.999. However, for the two higher concentrations, the fit remained unsatisfactory (R^2 = 0.874 and 0.895). The rate constant K decreased markedly with increasing C_0, from 1.284 to 0.025 L mg^{-1} min^{-1}. This trend suggests that at higher chlorine doses, the organic matter becomes the limiting factor in the reaction, reducing the apparent reactivity of chlorine [19-21]. Several authors have shown that K decreases as C_0 increases, and that this relationship depends on the organic content of the water (COD). This supports the idea that at higher chlorine doses, the organic matter becomes the limiting factor in the reaction [21].

The limited first-order model provided a good fit for all experiments, with R^2 ranging from 0.975 to 0.997, as shown in Table 4. The residual concentration C^* increased consistently with C_0 from 0.025 to 2.035 mg L^{-1}. This makes physical sense, as the organic matter concentration was constant in all experiments (oxidability = 4.6 mg O_2 L^{-1}). At higher chlorine doses, the available organic matter is insufficient to consume all the chloride, and a larger fraction therefore remains unreacted. The decay constant k was relatively stable across all experiments (0.302–0.393 min^{-1}), indicating that the reaction rate is largely controlled by the organic substrate rather than by the chlorine concentration.

The parallel first-order model splits chlorine decay into two phases: a fast initial reaction, described by k_1, and a slower phase involving less reactive compounds, described by k_2.

The parallel first-order model gave the best overall fit, with R^2 ranging from 0.985 to 0.999. The fraction x was approximately constant across all experiments (~0.4–0.5), meaning that approximately half of the initial chlorine reacted quickly and the other half reacted more slowly. This two-phase behaviour may reflect the presence of two groups of compounds with different reactivities. Either fast and slow reacting organic fractions, humic and fulvic compounds with different reactivities, where humic acids react faster with chlorine than fulvic acids [22]. Another possible explanation is the presence of inorganic reducing species such as Fe^{2+}, Mn^{2+}, NH_3, and NO_2^-, which react almost instantaneously with chlorine, followed by a slower reaction with organic matter. This second interpretation is particularly relevant for surface waters with a significant inorganic load, such as tropical river water.

Table 5 shows that k_1 was not statistically significant for the experiments with C_0 = 0.65 and 1.3 mg Cl_2 L^{-1}. The standard error exceeded the parameter value in both cases. A possible explanation is that the fast chlorine decay occurs within the first 3 minutes, and no measurements were taken in that period. This limits the ability of the model to estimate k_1 with confidence. For C_0 equal 2.7 and 4.7 mg L^{-1}, both constants were well determined, with k_1 clearly higher than k_2 (0.926 vs 0.019 min^{-1}; 0.444 vs 0.009 min^{-1}), confirming the two-phase decay behaviour.

The relationship between the parallel first-order model parameters and $1/C_0$ was also examined. Only k_2, showed a clear linear relationship with $1/C_0$ (R^2 = 0.997), described by Equation 5:

$$k_2 = 0.134\left(\frac{1}{C_0}\right) - 0.025 \quad (5)$$

For x and k_1, no clear trend was observed. This is largely because k_1 it was not statistically significant for the two lowest concentrations (C_0 = 0.65 and 1.3 mg Cl_2 L^{-1}), which limits any reliable analysis of its dependence on C_0. With only two valid estimates of k_1 (C_0 = 2.7 and 4.7 mg Cl_2 L^{-1}), no statistically meaningful conclusion can be drawn.

To establish these relationships, additional experiments at intermediate concentrations would be needed, with measurements taken within the first 3 minutes to ensure that $k_1$can be reliably estimated. This is suggested as a direction for future work.

Figure 3 (a–d) shows the experimental chlorine decay data for the initial concentrations tested, together with the fitted kinetic models.

To identify the model that best describes the experimental data, an *F-test* was applied to compare the four kinetic models. The F-test assesses whether the difference in fit between two models is statistically significant, or whether it can simply be attributed to random variation in the data.

The test is based on the ratio of the estimated variances of the two models being compared:

$$F = \frac{S_1^2}{S_2^2} \quad (6)$$

where $S_1^2$and S_2^2 are the estimated variances of the two models. If F exceeds the critical value, the model with the lower variance provides a statistically significantly better fit.

The degrees of freedom associated with each variance are calculated as $n - p$, where n is the number of experimental points (n n=7) and p is the number of model parameters. For the P1and P2 models, the degrees of freedom are 6. For PL model ($p = 2$) and PP model ($p = 3$), the degrees of freedom are 5 and 4, respectively.

All possible pairs of models were compared at a 95% confidence level. The degrees of freedom and the corresponding critical F values are summarised in Table 6.

Tab. 6. Degrees of freedom a critical F values (95%).

Comparison	df_1	df_2	F critical
P1 vs P2	6	6	4.28
P1 vs PL	6	5	4.95
PL vs P2	5	6	4.39
P1 vs PP	6	4	6.16
P2 vs PP	6	4	6.16
PL vs PP	5	4	6.26

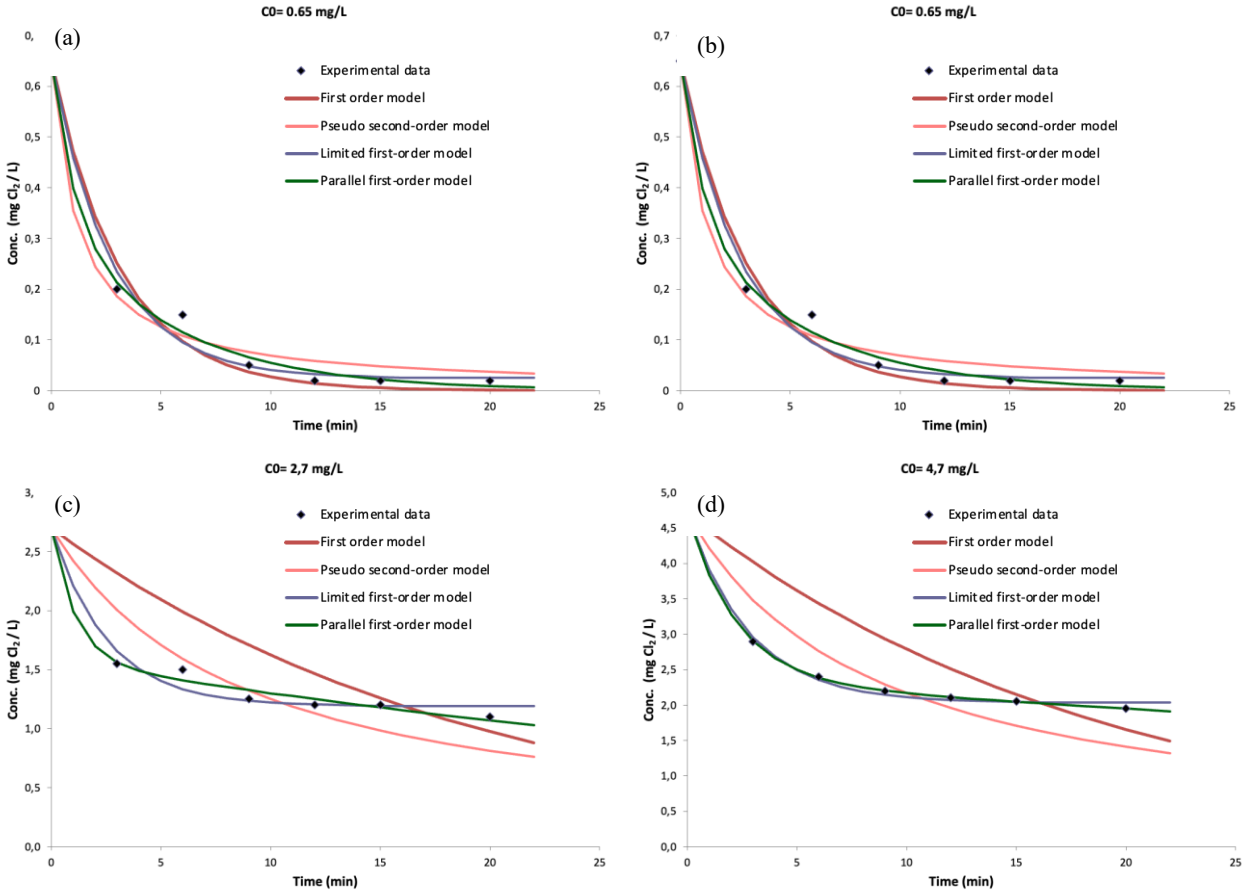

Fig. 3. Chlorine decay at T = 25 °C: (a) 0.65, (b) 1.3, (c) 2.7, and (d) 4.7 mg Cl_2 L^{-1} (initial concentration).

The F values obtained from the statistical comparison of the four models: first-order model (P1), pseudo second-order model (P2), limited first-order model (PL), and parallel first-order model (PP), are presented in Table 7.

Tab. 7. F-test at 95% confidence level for chlorine decay in water.

C_0 (mg Cl_2 L^{-1})	F calculated					
	P1/P2	P1/PL	PL/P2	P1/PP	P2/PP	PL/PP
0.65	1.26	0.948	1.335	1.182	0.934	1.248
1.3	1.43	1.36	1.059	2.236	1.558	1.650
2.7	1.13	2.11	0.534	5.195	4.596	2.455
4.7	1.16	2.04	0.567	107	92.3	52.3

For C_0 = 0.65 and 1.3 mg Cl_2 L^{-1}, the comparison involving the parallel first-order model (PP) was not performed, as k_1 was not statistically significant for these two conditions. For the remaining three models, all calculated F values were below the critical threshold, indicating that no model was statistically significantly better than the others (Tab. 7). However, F values for P1/P2 and PL/P2 were greater than 1, so P2 represents the best fit, confirming the earlier conclusions based on the R^2 values (0.99).

The *F-test* results for the third experiment (C_0 = 2.7 mg Cl_2 L^{-1}) showed no statistically significant difference between models. The largest F value was 5.195 (P1/PP), which did not exceed the critical value of 6.16. Nevertheless, all F values involving the PP model were greater than 1, and its R^2 was the highest, suggesting that it provided a consistently better fit, although not statistically significant at the 95% confidence level.

For C_0 = 4.7 mg Cl_2 L^{-1}, the parallel first-order model was statistically superior to all other models. The calculated F values for P1/PP, P2/PP, and PL/PP were 107, 92.3, and 52.3, respectively, all well above the critical values (6.16, 6.26). This result shows that the parallel model provides a significantly better description of chlorine decay at this concentration.

Overall, the parallel first-order model is the most appropriate for higher initial chlorine concentrations, where the two-phase decay behaviour is more pronounced. For lower concentrations, the simpler models are statistically equivalent and should be preferred.

3.2 Pratical application, contact tank design

The kinetic parameters obtained from the pseudo second-order model can be used directly to size a pre-chlorination contact tank. The contact time required to achieve a minimum residual chlorine concentration of 0.2 mg Cl_2 L^{-1} was calculated for each initial chlorine dose, using the expression derived from Equation (2).

For the Kifangondo Water Treatment Plant, with a flow rate of 138000 m^3 d^{-1} (95.8 m^3 min^{-1}), the required contact time and the corresponding tank volume were calculated for each initial concentration tested (Table 8).

Tab. 8. Contact time and tank volume for a minimum residual chlorine of 0.2 mg Cl_2 L^{-1} (Q = 138 000 m^3 d^{-1}, pseudo second-order model, plug flow assumption).

C_0 (mg Cl_2 L^{-1})	K (L mg^{-1} min^{-1})	T contact (min)	Tank volume (m^3)
0.65	1.284	2.7	258
1.3	0.289	14.6	1403
2.7	0.043	107.8	10342
4.7	0.025	193.0	18499

The results show that the initial chlorine dose has a strong influence on the required contact time and tank volume. A low dose (C_0 = 0.65 mg Cl_2 L^{-1}) requires only 2.7 min of contact and a small tank (~258 m^3). However, this dose offers little safety margin if water quality varies. A high dose (C_0 = 4.7 mg Cl_2 L^{-1}) requires 193 min and a tank of approximately 18500 m^3, which is neither practical nor economical, and would also increase the risk of trihalomethane formation. An intermediate dose of 1.3 mg Cl_2 L^{-1} appears to be the most balanced option, requiring approximately 15 min of contact and a tank volume of around 1400 m^3.

It should be noted that these calculations assume ideal plug flow conditions. In practice, real tanks have dead zones and hydraulic short-circuiting. A hydraulic efficiency factor (t_{10}/T) of 0.5 to 0.7 is typically applied, meaning the actual tank volume should be 1.4 to 2 times larger than the calculated value.

4. Conclusions

Chlorine decay in water occurs in two phases. The first is fast, involving reactions with more reactive species such as ammonia and certain organic compounds. The second is slower, involving less reactive compounds such as amines and humic substances.

Four kinetic models were tested to describe chlorine decay in raw water from the Bengo River at 25°C. The first-order model gave a poor fit at higher initial concentrations (R^2 = 0.72–0.74). The pseudo second-order model performed well only at lower doses. The limited first-order model gave a good fit across all conditions (R^2 = 0.975–0.997).

The parallel first-order model gave the best overall fit (R^2 = 0.985–0.999). At C_0 = 4.7 mg Cl_2 L^{-1}, it was statistically superior to all other models. For lower concentrations, no model was statistically superior to the others at the 95% confidence level.

For C_0 = 0.65 and 1.3 mg Cl_2 L^{-1}, the parameter k_1 was not statistically significant, likely due to the absence of measurements in the first 3 minutes. Shorter sampling intervals are recommended in future work.

Finally, the kinetic parameters can be used directly to size pre-chlorination contact tanks. The required tank volume was shown to vary significantly with the initial chlorine dose.

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