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Wastewater disinfection using sodium dichloroisocyanate (NaDCC) and sodium hypochlorite (NaOCL): Modeling, optimization and comparative analysis

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ABSTRACT

The aim of this study was to evaluate the ability of sodium dichloroisocyanate (NaDCC) and sodium hypochlorite (NaOCL) for wastewater disinfection in a batch system with considering the contact time (5, 10, 15 min) and disinfectant concentration (0.01, 0.02, 0.04, 0.06, 0.08, 0.1, 0.2, 0.4 mg/L). Distilled water was mixed with wastewater to produce a given concentration of bacteria. Pure plate count (PPC) method was used to determine the initial and final concentration of bacteria. Three regression models including first order (FO), first order plus two way interaction (FO + TWI), and second order (SO) were used to fit the experimental data. The differences among group means were analyzed using Kruskal–Wallis and ANOVA analysis. The removal efficiency of NaDCC and NaOCl were 88.7 and 52.38% at an initial concentration of 0.01 mg/L and contact time of 10 min. The results of Kruskal–Wallis test showed that the mean of disinfection efficiencies were statistically different at different concentration of disinfectant (p-value < 0.05). However, there was no statistically significant difference between removal efficiency at different contact time based on one way ANOVA analysis (p-value > 0.05). These results were the same for both disinfectants. The modeling data showed that the best regression model to describe the disinfection mechanism was SO model with R^2 value of about 0.84 for both NaDCC and NaOCl. According to the present study, it can be concluded that the NaDCC is more suitable for water and wastewater disinfection because of higher efficiency at lower contact time and concentration.

Keywords: Sodium dichloroisocyanate (NaDCC); Sodium hypochlorite (NaOCl); Wastewater; Disinfection; Modeling

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1. Introduction

Over the past 100 years, chlorine and its compounds have been used as the most common and acceptable agents for water and wastewater disinfection and sanitation worldwide. The most important advantages of chlorine compounds are their low cost, high germicidal power, and relatively long durability [1–3]. The most applicable forms of the chlorine in the water and wastewater treatment are elemental chlorine (Cl_2), calcium hypochlorite $Ca(OCl)_2$, sodium hypochlorite (NaOCl), lithium hypochlorite (LiOCl) and chloroisocyanorutes (sodium dichloroisocyanurate and trichloroisocyanuric acid) [4]. Sodium hypochlorite (NaOCl) is available in the liquid form with 5–15% free chlorine. The main advantages of NaOCl are non-toxicity, high safety, rapid penetration into cellular membranes [5], and evident antimicrobial activity [6,7]. However, there are some limitations with this compound which are high reactivity and instability, and high sensitivity to light and high temperatures. Also, after a month, its primary disinfection power and reactivity is reduced to one-third. The other disadvantages of this compound are high corrosivity, skin irritation, and storage problems [1,4,8]. Sodium dichloroisocyanurate (NaDCC) is one of the chlorine compounds that can be used as substitute for NaOCl [9]. NaDCC with commercial names of troclosence sodium and dichloro-s-triazinetrione is in the form of powder and tablet and has 62% available chlorine. The most important advantages of NaDCC in comparison with NaOCl are its lower cost, higher stability, safety, and solubility, easier handling, measurement, transportation, and storage, higher shelf life, more precise dosage, and lower chemical risk and by-products formation probability [10,11]. NaDCC already is widely used for water disinfection in emergency situations. In addition to, NaDCC has been widely used as tablets in the home scale for drinking water disinfection and has been shown a good performance in the fecal coliform removal [12,13]. This compound has recently approved by the EPA and WHO for conventional treatment of drinking water [4,14,15]. Beside, NaDCC has been applied for the disinfection of the swimming pool, industrial cooling tower, and hospital, and the sterilization of baby milk bottles [16]. Several studies have been conducted to evaluate germicidal effect of NaDCC. Ayorya et al. (1998) published a paper in which they demonstrated that NaDCC can effectively kill gram-positive, gram-negative, and fungi [17,18]. The disinfection efficiency of NaDCC and NaOCl on mesophilic aerobic bacteria, fungi, yeast, total coliforms, and salmonella was studied by Nascimento and colleagues. They found that NaDCC in the same concentration as NaOCl was more effective for killing the mentioned microorganisms [11]. In another study it was shown that NaDCC at the same concentration is more effective than NaOCl against Vibrio cholera [19]. In 1981, Mazola et al. demonstrated that NaDCC is more effective than NaOCl for disinfection of hospital rooms and instruments. They concluded that higher disinfection efficiency by NaDCC may be originated from higher germicidal power, slower decomposition and HOCl releasing in the longer time, higher capacity for maintaining available free chlorine, and more resistance to pH changing [20,21]. Also, a number of studies have been conducted for assessing effect of NaDCC on *Streptococcus sobrinus*, *Streptococcus salivarius*, *Enterococcus faecalis*, *Streptococcus mutans* [22], *Clostridium difficile* spores [23], and *Botrytis cinerea* [24]. So far, however, there has been a little discussion about the impact of NaDCC on gram-negative bacteria. Therefore, this paper will focus on the study of NaOCl and NaDCC as a disinfectant for the removal of gram-negative bacteria from the aqueous solution.

2. Material and methods

2.1. Chemicals and solutions preparation

The chemical reagents used for this study, such as sodium hypochlorite (NaOCl), sodium dichloroisocyanurate (NaDCC), Eosin methylene blue agar (EMB), and acid nitric were provided from Merck Company, Germany. All chemicals were analytical grade. All glassware was washed with acid nitric and then rinsed three times with distilled water.

A stock solution of NaDCC (1500 mg/L) was prepared by dissolving accurate quantities (1.5 g) of standard powder in 1 l double distilled water. The 5% (50000 mg/L) commercial solution of NaOCl was used as a stock solution. The working standard solutions (0.01, 0.02, 0.04, 0.06, 0.08, 0.1, 0.4, 0.8, 1, 2, 10 mg/L) of NaDCC and NaOCl were prepared from the stock solution by serial dilution.

2.2. Disinfection experiments

The batch experiments were conducted to determine the efficiency of NaOCl and NaDCC to remove gram-negative bacteria. The disinfection experiments were performed in 100 ml glass beaker as reaction reactor in which microorganism concentration, disinfectant concentration, and contact time variations were managed. The specific volume of stock solution was transferred to the beaker and diluted with wastewater to produce an intended solution with a given concentration of microorganisms. After this, at different interval times (5, 10, and 15 min) the samples were taken and the concentration of gram-negative bacteria was determined using pure plate count (PPC) method. For this aim, after a given period of time, 1.0 ml of the final solution (combined wastewater and disinfectant solution) was transferred to a plate and two-thirds of the plate was poured with eosin methylene blue agar (EMB). After cooling, the plate was placed upside down at 35 ± 5°C for 24 h in an incubator. Finally, the bacteria were counted using colony counter. The sampling and testing were conducted according to Standard Methods for the Examination of Water and Wastewater [25].

2.3. Modeling and optimization

Modeling of the removal efficiency was conducted using RSM package in R software version 3.1.3 [26]. Three models, including first order (FO), first order plus two way interaction (FO + TWI) and second order (SO) or full quadratic were fitted to the data.

In the FO model, only the effect of input variables (concentration and time) on removal efficiency was measured in linear terms. The first order model is shown as the following equation [27,28]:

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \varepsilon \quad (1)$$

where k is the number of input variables, β_0 is the constant term, β_i represents the coefficients of the linear variables, x_i represents the input variables, and ε is the error associated with the experiments.

In the FO + TWI model, beside the linearity effect, interactive impacts of concentration and time on removal efficiency were evaluated. The second-order model is described using the following equation [27]:

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{1 \le i \le j}^{k} \beta_{ij} x_i x_j + \varepsilon \quad (2)$$

where β_{ii} represents the interaction coefficients between input variables.

The last and most complicated model was quadratic model that contains characteristic of the two previous models plus polynomial function. The quadratic model is described using following equation [27,29]:

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{1 \le i \le j}^{k} \beta_{ij} x_i x_j + \varepsilon \quad (3)$$

where β_{ii} represents the coefficients of the quadratic parameter.

The quality of the polynomial model fitness was expressed by the multiple R square, adjusted R square and lack of fit. Model terms were evaluated by the p-value (probability) at the 0.05 level. The quadratic model with linear term, interaction term and square term was used for predicting the optimal conditions. Optimization was performed using solver add-ins function in Microsoft Excel 2013.

3. Results and discussion

3.1. Impact of disinfectant concentration

The impact of contact time on removal efficiency were shown in Figs. 1 and 2. As can be seen from these figures, for both disinfectants (NaOCl and NaDCC), the disinfection efficiency was increased as the disinfectant concentration was increased. The efficiency of NaDCC was 88.7% for the concentration of 0.01 mg/L while it was 100% for 0.4 mg/L at contact time of 10 min. For NaOCl, increasing in disinfectant concentration from 0.01 to 0.4 mg/L led to increasing in disinfection efficiency from 52.38% to 100%.

The results of Kruskal–Wallis analysis showed that there is a statistically significant difference between the averages of disinfection efficiencies at different disinfectant concentrations for both disinfectants ($P < 0.05$). Multiple comparison test after Kruskal-Wallis showed that for NaOCl, average of efficiencies at concentrations of 0.01–0.02, 0.01–0.06, and 0.02–0.06 and for NaDCC at concentrations of 0.01–0.1, 0.01–0.2, and 0.01–0.4 were statistically different.

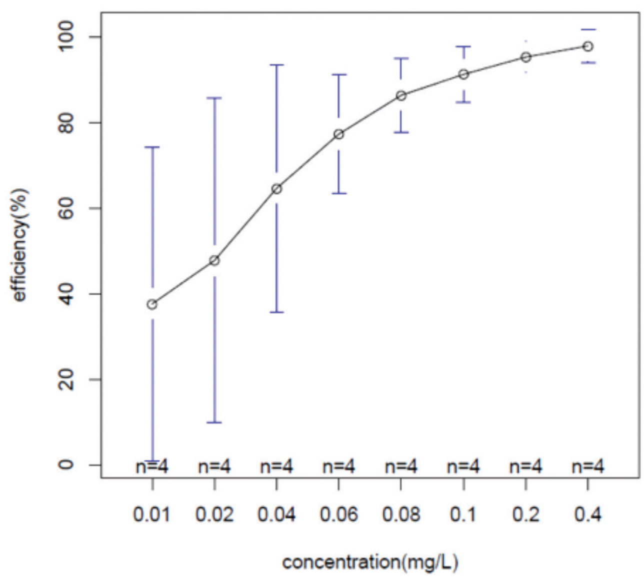

Fig. 1. Mean plot of disinfection efficiency at different concentrations of NaOCl.

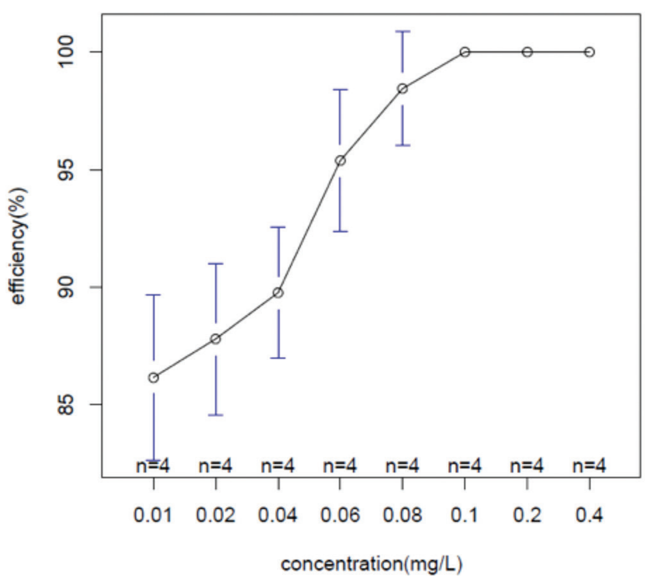

Fig. 2. Mean plot of disinfection efficiency at different concentrations of NaDCC.

3.2. Impact of contact time

The mean plots of removal efficiencies at different contact times were shown in Figs. 3 and 4. It can be seen from the figures that disinfection efficiencies of disinfectants were increases as the contact time were increased. NaOCl results show that by increasing the contact time from 5 to 15 min, removal efficiency was increased from 80.1% to 93.3%. For NaDCC, removal efficiency was increased from 96.4% to 100% when contact time was increased from 5 to 15 min.

The results of one-way ANOVA analysis showed that there is no statistically significant difference between the averages of disinfection efficiencies at different contact times for both disinfectants ($P > 0.05$).

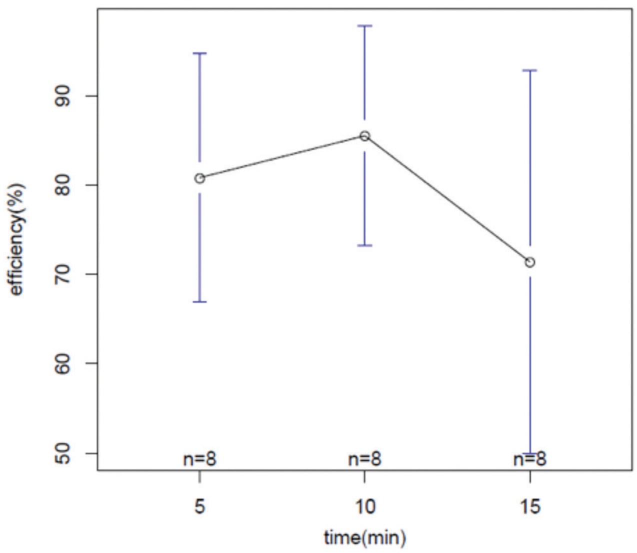

Fig. 3. Mean plot of disinfection efficiency of NaOCl at different contact times.

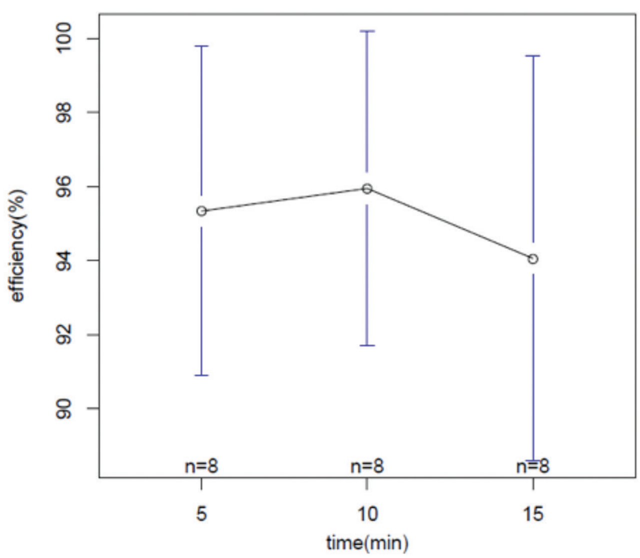

Fig. 4. Mean plot of disinfection efficiency of NaDCC at different contact times.

3.3. Models analysis

3.3.1. FO model

The results of the FO model that was fitted to the data were shown in Table 1. As can be seen from the table, the effect of concentration on the disinfection efficiency of NaOCl and the intercept of the model is statistically significant at the 0.001 level. But, the effect of time is not significant. The obtained model is according to the following equation.

$$\text{Removal efficiency} = 89.97478 + 28.58434\text{C} + 0.18900\text{T} \quad (4)$$

The obtained FO model is significant at the 0.01 level. For NaDCC, the effect of concentration, contact time and intercept is significant at the 0.001, 0.1 and 0.001 levels, respectively. The mathematical FO model for NaDCC is as follows:

$$\text{Removal efficiency} = 53.29649 + 104.23963\text{C} + 1.41012\text{T} \quad (5)$$

This model is significant at the 0.001 level.

3.3.2. FO + TWI model

The results of the FO + TWI model are shown in Table 2. The results show that for NaOCl, the intercept and concentration are significant at the 0.001 and 0.1 levels, respectively. However, the impact of contact time and interactive impact of concentration and contact time on removal efficiency are not significant. The mathematical FO + TWI model for NaOCl is as follows:

$$\text{Removal efficiency} = 88.86 + 38.34928\text{C} + 0.3\text{T} - 0.9764\text{CT} \quad (6)$$

The model is significant at the 0.01 level.

For NaDCC, the impact of concentration, contact time and intercept are significant at the 0.05, 0.05 and 0.001 levels, respectively. The interactive impact of concentration and contact time is not significant. The mathematical FO + TWI model for NaDCC is as follows:

$$\text{Removal efficiency} = 46.66 + 162.53C + 2.0732T - 5.82931CT \quad (7)$$

The obtained model is significant at the 0.01 level.

3.3.3. SO or quadratic model

The results of the first-order effects (C and T), interaction effect (CT) and second-order effects (C^2 and T^2) for quadratic model are shown in Table 3. The results show that for NaOCl, the intercept, first order, and second order effects of concentration on disinfection efficiency are significant at the 0.001 level. The effects of the other terms on disinfection efficiency are not statistically significant. The quadratic model for NaOCl is as follows:

$$y = 82.87 + 144.6\text{C} + 0.567\text{T} - 0.9764\text{CT} - 259.61\text{C}^2 - 0.0133\text{T}^2 \quad (8)$$

The model is significant at the 0.001 level.

For NaDCC, the first and second order effects of concentration on disinfection efficiency are statistically significant at the 0.001 level. The effect of other parameters on removal efficiency is not statistically significant. The mathematical SO model for NaDCC is as follows:

$$y = 23.61 + 492.4x + 3.96z - 5.83xz - 805.9x^2 - 0.0942z^2 \quad (9)$$

The final quadratic model for NaDCC is significant at the 0.001 level.

Table 1
Parameters and its coefficient in terms of FO model for NaOCl and NaDCC

Disinfectant type	Parameters	Intercept	C	T
NaOCl	Coefficient	89.97478	28.58434	0.18900
	P-value	2.2 (10–16) ***[1]	0.0007054***	0.3892413
	Model *p*-value	0.002271		
NaDCC	Coefficient	53.29649	104.23963	1.41012
	P-value	1.661 (10–6) ***	0.0002561***	0.0596521
	Model *p*-value	0.0004011		

[1]Significant codes: ***(0.001), **(0.01), *(0.05), · (0.1).

Table 2
Parameters and its coefficient in terms of FO + TWI model for NaOCl and NaDCC

Disinfectant type	Parameters	Intercept	C	T	C:T
NaOCl	Coefficient	88.86402	38.34928	0.30008	–0.97649
	P-value	2(10–16)***[1]	0.06201·	0.32796	0.59265
	Model *p*-value	0.006837			
NaDCC	Coefficient	46.66565	162.53276	2.07321	–5.82931
	P-value	0.0002414***	0.0176145*	0.0449408*	0.3282256
	Model *p*-value	0.001012			

[1]Significant codes: ***(0.001), **(0.01), *(0.05), · (0.1).

Table 3
Parameters and its coefficient in terms of SO or quadratic model for NaOCl and NaDCC

Disinfectant type	Parameters	Intercept	C	T	C:T	C^2	T^2
NaOCl	coefficient	82.872126	144.608432	0.5670	–0.9764	–259.61	–0.0133
	P-value	9.923e-14***[1]	8.718e-07***	0.5315	0.3574	4.107e-06***	0.7630
	Model *p*-value	1.718e-06					
NaDCC	coefficient	23.661791	492.404071	3.958709	–5.8293	–805.952	–0.0942
	P-value	0.1212	1.290e-06***	0.2200	0.1251	1.855e-05***	0.5450
	Model *p*-value	1.195e-06					

[1]Significant codes: ***(0.001), **(0.01), *(0.05), · (0.1).

3.3.4. Models comparison

The *F*-test lack of fit demonstrates the variation of the data around the fitted model. The small *P* values for lack of fit (less than 0.05) show that the model does not fit the data well. The lack of fit of all models for both disinfectants is not significant. These results suggest that all the models are suitable in terms of fit to the experimental data.

The multiple R^2 coefficient shows the total variation in the dependent factor predicted by the model. A higher multiple R^2 value, close to 1, shows lower variation of the response to the experimental. For desirable condition, a reasonable agreement between multiple R^2 and adjusted R^2 is necessary. As can be seen from Table 4, the obtained quadratic model for both NaOCl and NaDCC has a higher multiple and adjusted R^2 rather than first order and two way interaction model. Also, the difference between two R^2 is reasonably low. So, the quadratic model was selected as the best fitted model for optimization and plotting.

The quadratic model plots for NaOCl and NaDCC based on the interactive effect of the two factors (contact time and concentration) are shown in Figs. 5 and 6.

3.4. Optimization

The optimal condition shows the best set of independent parameters (concentration and contact time) to acquire maximum disinfection efficiency. The results of optimization show that maximum efficiency 100% can be obtained at the contact time of 10 min and the concentration of 0.125 mg/L for NaOCl and 10 min and 0.144 mg/L for NaDCC.

Table 4
Comparative analysis of first order, two way interaction and quadratic models

Disinfectant type	Model's parameters	First order	Two way interaction	Quadratic
NaOCl	Multiple *R*-squared	0.44	0.4481	0.8354
	Adjusted *R*-squared	0.3866	0.3654	0.7897
	Lack of fit	18.484	19.126	6.338
NaDCC	Multiple *R*-squared	0.5252	0.5479	0.8421
	Adjusted *R*-squared	0.48	0.4801	0.8923
	Lack of fit	200.65	200.61	0.7784

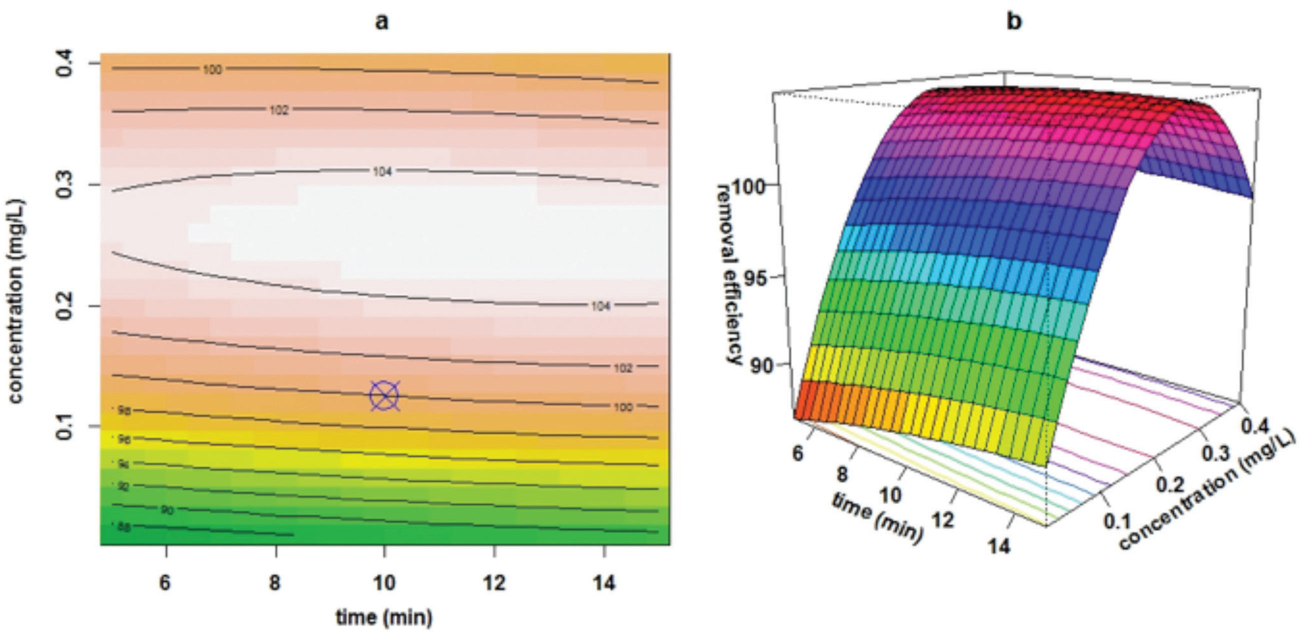

Fig. 5. Contour (a) for respective three dimensional curve, and (b) for interactive effect of time and concentration on disinfection efficiency of NaOCl.

4. Conclusion

On the basis of the results, both NADCC and NaOCl are efficient disinfectant for killing gram-negative bacteria. The removal efficiencies obtained in some initial disinfectant concentration of NADCC is higher than NaOCl. This can be due to the fact that NaOCl release all the free available chlorine in an initial time of disinfection. However, NaDCC release almost half of the available chlorine at the first moment of the contact time and the residual disinfectant is stored as chlorinated isocyanurate [30,31]. Also, this fact can be justified with decreasing pH when NaDCC is applied as disinfectant and increasing pH when NaOCl is used. The results of this research in terms of higher antimicrobial power of NaDCC rather than NaOCL are consistent with the findings of other studies [17,21,22]. The quadratic model is the best model to describe the results of disinfection using NaOCl and NaDCC. This fact shows that interactive effect of contact time and concentration has an important role in disinfection efficiency.

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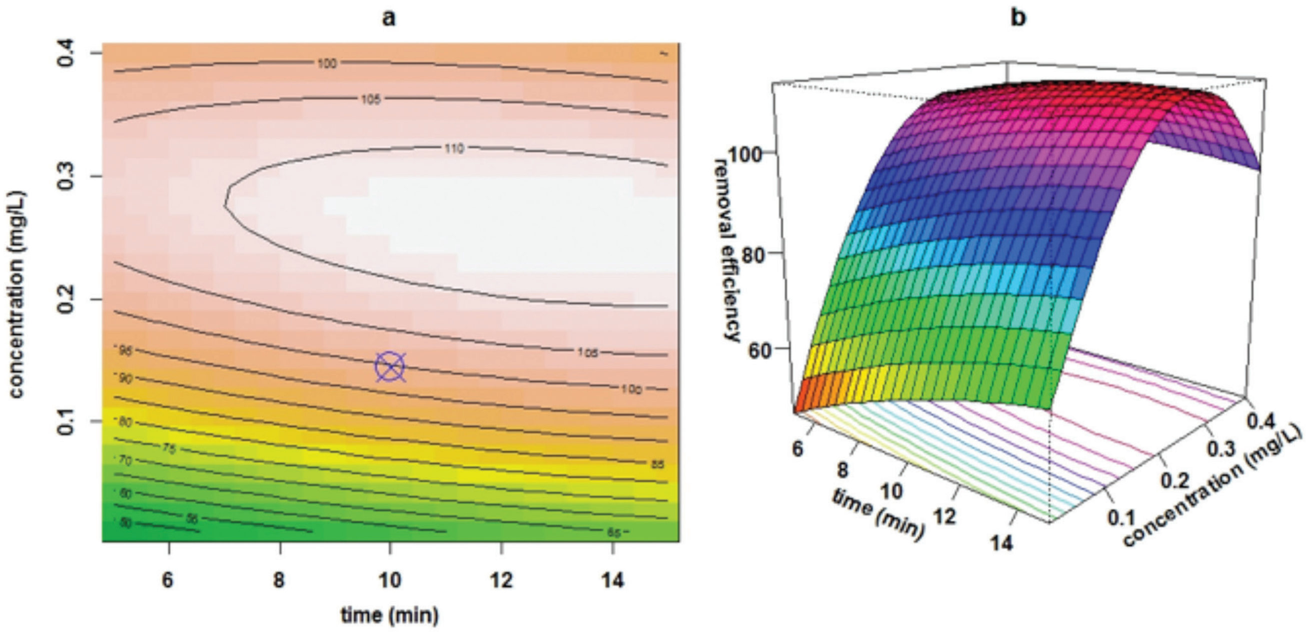

Fig. 6. Contour (a): for respective three dimensional curve and (b): for interactive effect of time and concentration on disinfection efficiency of NaDCC.

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