Journal of Research in Science Engineering and Technology
Volume 9, Issue 1, December 2021, pp. 38-49

Journal of Research in Science Engineering and Technology

www.researchub.org

Smart Controlling Cyanide Emissions from Surface Water Resources by Predictive Models: An integrated GA-Regression

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Abstract
Maintaining water resources safety is a matter of importance in sustainable urban management, which would directly influence the quality of drinking water. Surface water resources have high risk of contamination with hazardous substances through intentional or unintentional factors. The present study attempts to investigate and provide strategies to remove cyanide contamination, which has resulted from permeation or subversive injection, with chlorine for a water treatment plant in Iran. In this study contamination has been defined in three scenarios including low ([CN-] = 2.5 mg L-1), medium ([CN-] = 5 mg L-1), and high ([CN-] = 7.5 mg L-1) levels, and optimal doses of injected chlorine have been suggested as 2.9 mg L-1 (low contamination), 4.7 mg L-1 (medium contamination), and 6.1 mg L-1 (high contamination), respectively. Finally, the Gaussian distribution, calibrated through genetic algorithm, has been presented as the best model for determining the residual amount of cyanide according to the injected chlorine.

Keywords
Chlorine, Cyanide, Genetic Algorithm, Sabotage Operations, Water Treatment

1. Introduction
Maintaining water safety is considered as one of the major responsibilities of water resources management system (Fathollahi-Fard et al., 2020a). Developing countries or even the developed ones require such maintenance of water safety to reach full realization of sustainable development. Consequently, preserving qualitative and quantitative parameters of water during any process such as supplying resources, treatment, transfer and distribution, is of utmost importance. There are different scenarios for water contamination which, in general, are classified into executive (intentional) and non-executive (unintentional) factors (Fathollahi-Fard et al., 2020b). A historical investigation of threatening phenomena

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Received 26 September 2020 / Accepted 15 November
DOI: https://doi.org/10.24200/jrset.vol9iss01pp38-49

concerning water resources shows that changes in the quality of water, either through intentional or unintentional events, poses a higher risk towards the consumers' health. Developing countries with lack of engineered environmental facilities including landfills, wastewater treatment plants, and the industrial treatment units, in particular, provide the possibility of contamination leakage to both surface and underground water resources. Also, the occurrence of various acts of terrorism, especially in the Middle East, is regarded as a potential threat towards water resources. Threats concerning water are divided into three classes of physical, chemical, and biological phenomena in which the damages caused by the last two cases are far more severe than the latter, i.e. physical criteria (Bradley et al., 2006; Vörösmarty et al., 2010). When a network is contaminated with a chemical factor, four aspects including detection time, source of contamination, intensity of contamination, and the elapsed time since its commencement, would gain significance concerning how to provide a proper management and engineering response.

Studies on intentional or unintentional entry of contaminants in water resources can be divided into two classes of network contaminations and contaminations prior to network. Among the studies on the safety of water distribution networks, a research conducted by Preis & Ostfeld (2008) can be taken into consideration. In this study, by using non-dominated sorted genetic algorithm analysis (NSGAII), it has been attempted to locate sensor placements in distribution network. In another study conducted by Krause et al. (2008), such determination of sensor placements has been carried out in large networks. Investigations in this study have shown that using the Mixed Integer Programming (MIP) method does not involve adequate performance. Therefore, algorithms based on multi-criteria decision analysis have been applied (Krause et al., 2008). Nicklow et al. (2010) have examined and compared two engineering and algorithmic design strategies in locating water distribution sensor placements. With regards to various acts of terrorism and in order to locate water system sensors, this study emphasized on integration of both the algorithmic and engineering approaches (Nicklow et al., 2010). Arad et al. (2013) have designed a dynamic system for identifying the contamination in water distribution systems. This system is defined as online and offline modes; in which online and offline decision variables are adjusted through recursive Bayes rule and GA (Genetic Algorithm), respectively. Zhao et al. (2016) have also determined the best location for placing contamination detection sensors in water networks using Kosari optimization technique.

Cyanide as a lethal contaminant is a critical criteria in maintaining water safety (Ware et al., 2006). Affordability, ease of access, quick impact, effectiveness even at low concentrations ($[CN^-] > 70$ μg L^{-1}), no physical traces (like color, odor or turbidity) and the possibility of contamination by industries such as metal plating makes cyanide a potential terroristic threat (Dzombak et al., 2005).

Several researches were conducted with the aim of removing cyanide during water treatment process (Hijosa-Valsero et al., 2013; Pirmoradi et al., 2017; Alıcılar et al., 2002). Dash et al. (2009) focused on the ability of anaerobic microorganisms in decomposition of cyanide. In fact, such microorganisms would transform the carbon and nitrogen in cyanide to carbonate and ammoniac, respectively. The above-mentioned approach is possible for the treatment of cyanide from industrial wastewater which has the potential for anaerobic microorganism to grow (Dash et al., 2009). Parga et al. (2003) have also attempted to treat the cyanide waste solutions by employing three techniques including (1) oxidation by chlorine oxide (ClO_2) in gas-sparged hydrocyclone reactor (GSH) system, (2) ozonation in batch reactors with additional intense shaking (Stirred Batch Reactor), and (3) UV light. All of the applied methods successfully removed cyanide, each with different advantageous aspects (Parga et al., 2003). Another research carried out in the area of the adverse effects of cyanide on human health, examined different treatment procedures for removal of cyanide from industrial wastewater (Arbabi et al., 2015). In this research, the performance of various treatment processes such as chlorination, biological treatment, acid removal, evaporation, ion exchange, oxidation with hydrogen peroxide, etc. for removal of cyanide were tested. Uppal et al. (2017)

attempted to remove cyanide from water resources using the zinc peroxide nanoparticles (ZnO2) along with PVP (polyvinyl pyrrolidone) stabilizing factor based on surface adsorption process. This process mainly depends on the pH, concentration of adsorption material (ZnO2-PVP), contact time and concentration of cyanide.

The aim of the present research is to determine the optimal doses of injected chlorine for the removal of cyanide contamination. For this purpose, three contamination scenarios of cyanide for a known water treatment plant in Iran have been assessed. Finally, with the aim of developing smart models for encountering with cyanide contaminations, a set of mathematical relationships have been obtained for addressing the residual amount of cyanide with regards to the concentrations of injected chlorine.

The rest of this paper is followed by three sections. Section 2 studies the materials and methods of this research. Section 3 does the computational results and analyses. Finally, Section 4 concludes this research and future suggestions.

2. Materials and Methods

Security protocols do not allow us to present the case study description. Field survey assessments have demonstrated the fact that there are a number of industrial factories in the vicinity of this WTP such as metal plating and tubing. Considering nonconformities towards maintaining environmental standards in treatment of industrial wastewater, the leakage of cyanide into the drinking water supplies is probable. On the other hand, the water flow between the dam and the WTP is not transferred via channel or a pipeline and flows gravitationally along the river bed. Such conditions may intensify the possibility of industrial wastewater leakage or facilitate various sabotage operations. The studied WTP applies chlorination in two stages of primary and final in order to carry out the disinfection process. Samples in this study have been collected from the water entering WTP.

2.1. Determining the Optimal Concentrations of Chlorine in Different Scenarios

In this part of the study, three scenarios involving low (2.5 mg L-1), medium (5 mg L-1), and high cyanide contamination (7.5 mg L-1) have been defined. During each experiment, the samples were contaminated by the certain concentrations of cyanide and then different doses of NaOCl injected into the samples and the residual cyanide were detected.

The United States Environmental Protection Agency (USEPA) considers the maximum contaminant level for free cyanide in surface water resources to less than 200 μg L-1 (USEPA, 2009). Meanwhile, the 1053 standard in Iran states the limit for existing cyanide in water supplies as 70 μg L-1 (DOE, 2016). Nonetheless, the present study carries out the cyanide removal process until the concentration of cyanide reached to the 70 μg L-1 in order to satisfy the current condition of standard in Iran.

To measure the residual amount of cyanide in the chlorinated sample, the present study employs a patented method known as US 4871681A which is demonstrated in Table 1 (Bilger & Wolf, 1989). Also, in order to record the absorbance of the sample solutions, Agilent 8453 spectrophotometer equipped with photodiode array detector was used.

Table 1. Stages of cyanide detection experiment according to US Patent 4871681A (Bilger & Wolf, 1989)

Test Stages	Description
1	Pour 25 mL of the testing solution in the beaker
2	Add 5 mL of Na_2CO_3 0.5 mol L^{-1}

3	Add 5 mL Picric acid (1%w/v) into the beaker
4	Heat the container to near boiling point to get the colour changes
5	Let the samples to cool at room temperature
6	Measure the absorptions of the standard and testing samples at the wavelength of 520 nm

2.2. Reagents

Deionized water was used throughout the analysis. A stock solution of 0.5 mol L-1 Na2CO3 was prepared from Na2CO3 salt (Merck, Darmstadt, Germany). A stock solution of NaOCl 5 mol L-1 was prepared from Merck, Darmstadt, Germany and standardized according to the 4500-CI. B Iodometric Method I (APHA, 2005). A solution of 1% (w/v) of Picric acid (Merck Company) was used as a reagent for determination of cyanide. A stock solution of 1000 mg L-1 CN- was prepared from KCN salt (Merck, Darmstadt, Germany).

2.3. Modelling the Residual Cyanide (RCN)

Mathematical modelling of the experimental data has been carried out with the purpose of providing a smart relationship between concentration of cyanide and the required dosage of chlorine for the removal of contamination. For this purpose, a set of mathematical distributions including Polynomial, Exponential, Fourier, Gaussian, and Rational have been used. The appropriate distribution has been chosen through evaluation of statistical parameters of R2, SSE and RMSE indices.

Prior to the modeling, it is required to interpolate the contour between values of injected chlorine and RCN output using Lagrange method which expressed in Eq. 1. It is worth mentioning that all the interpolation calculations as well as the mentioned modelling procedures have been carried out in MATLAB 2015a software.

$$f\left(x_k\right)=P\left(x_k\right),\ for\,each\,k \cong 0,1,2,\ldots n \tag{1}$$

$$P\left(x\right)=\sum_{k=0}^{n} f\left(x_k\right) L_{n,k}\left(x\right)$$

$$L_{n,k}\left(x\right)=\prod_{\substack{i=0 \\ i\neq k}}^{n} \frac{\left(x-x_i\right)}{\left(x_k-x_i\right)}$$

2.4. Calibration of Model Using Genetic Algorithm (GA)

Subsequent to determining the relationships between the concentration of injected chlorine and residual cyanide in water, the obtained relationships are adjusted using model calibration tests along with single-purpose genetic algorithm. In fact, theoretical (calculated through predictive models) and practical values (experimental results) are compared in accordance with the cost function equation presented in Eq. 2. By using this method, the coefficients of the proposed models are calibrated with the aim of lowering the cost function (Eq. 2).

$$Cost\ \ function = min(R_e - R_t)^2 \tag{2}$$

$$R_e = experimental\ response$$

R_t=Theoretical Response (in polynomial model, $R_t = a_0 + a_1 C_{Naocl} + a_2 C_{Naocl}^{2} + \ldots a_n C_{Naocl}^{n-1}$)

Result of equation Determine $a_0, a_1, \ldots a_n$

Rt=Theoretical Response (in polynomial model,)

3. Result of Equation

To analyze the abovementioned issue, it should be noted that the GA algorithm operation has been coded in MATLAB 2015a software. According to the research study conducted by De Jong, set parameters of mutation rate, crossover probability, and initial population were considered as 0.001, 0.6 and 50, respectively (De Jong, 1975). Sensitivity analysis of algorithm's behavior concluded the end value of 400 generations.

3.1. Results and Discussion

As mentioned before, the studied WTP used chlorine in its gas state. Meanwhile, the results obtained from the studies by Botz have shown that the final product of reaction between Chlorine gas (Cl2) and cyanide ion does not produce dangerous components according to Eq. 3 (Botz, 2001).

$$Cl_2 + CN^- \rightarrow CNCl + Cl^- \tag{3}$$

$$CNCl + H_2O \rightarrow OCN^- + Cl^- + 2H^+$$

$$OCN^- + 3H_2O \xrightarrow{Cl_2\ catalyst} NH_4^+ + HCO_3^- + OH^-$$

$$3Cl_2 + 2NH_4^+ \rightarrow N_2 + 6Cl^- + 8H^+$$

In this study, the optimal concentrations of chlorine for the removal of cyanide in low (2.5 mg L-1), medium (5 mg L-1) and high (7.5 mg L-1) contamination scenarios have been calculated as 2.9, 4.7, and 6.1 mg L-1, respectively. The relationship between the injected chlorine and the residual cyanide in low, medium and high degrees of contamination were illustrated in Figures 1, 2, and 3, respectively. As it can be seen, by increasing the concentration of injected chlorine, the residual cyanide is decreased non-linearly. Noticeably, due to the high concentrations of volatile solids (VS) in surface waters, chlorination with the obtained optimal doses increases the possibility of Trihalomethane formation drastically. For solving this problem, two strategies including initial disinfection by using potassium permanganate and multi-stage chlorination are recommended. Due to the color formation by using high concentrations of potassium permanganate (higher than 1 mg L-1), its application encountered many limits. Therefore multistage chlorination is preferred.

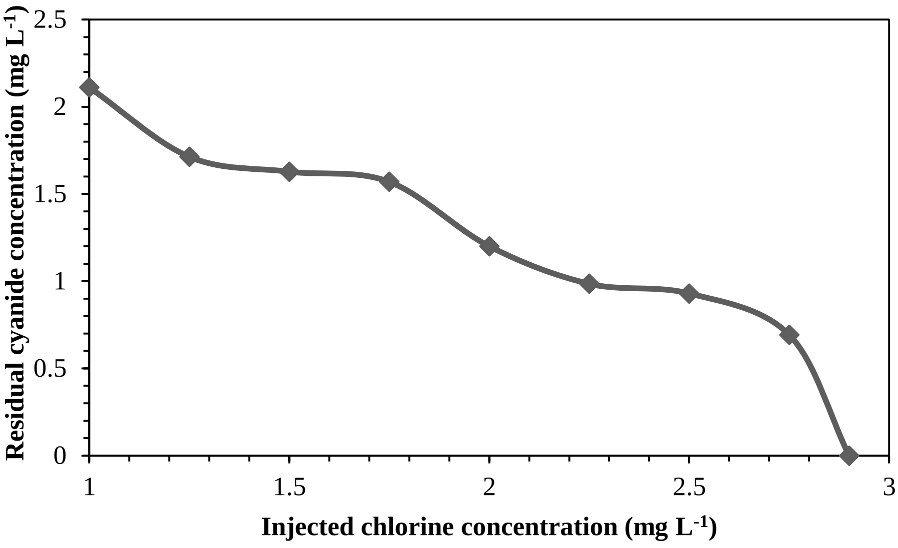

Fig. 1. Diagram of changes of the residual cyanide in water at various concentrations of the injected chlorine – low contamination scenario ([CN-] = 2.5 mg L-1)

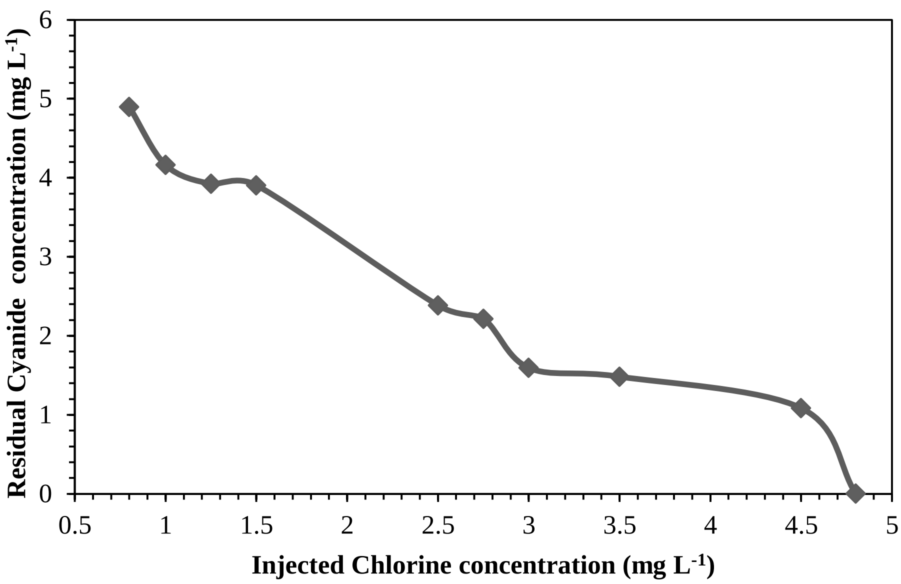

Fig. 2. Diagram of changes of the residual cyanide in water at various concentrations of the injected chlorine – medium contamination scenario ([CN-] = 5 mg L-1)

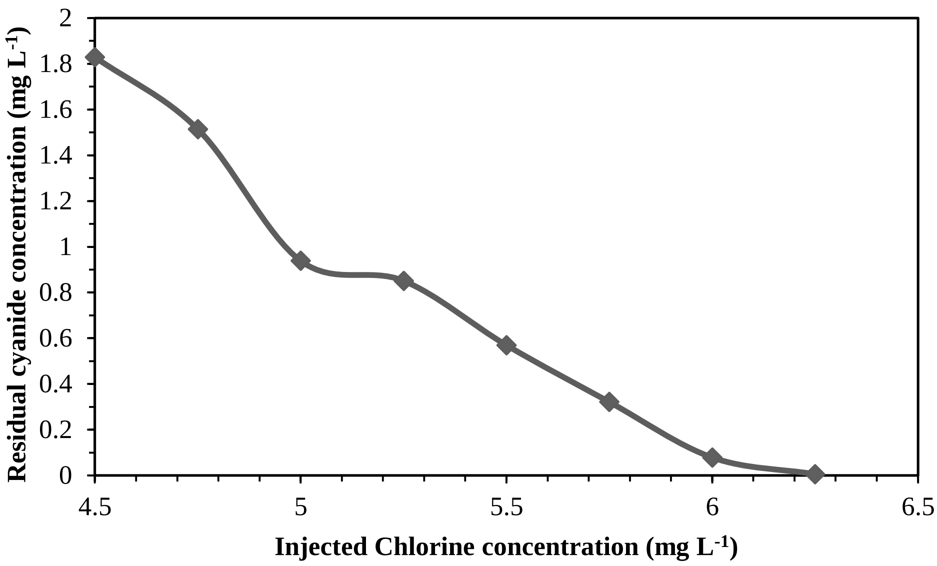

Fig. 3. Diagram of changes of the residual cyanide in water at various concentrations of the injected chlorine – high contamination scenario ([CN-] = 7.5 mg L-1)

With the purpose of predicting residual cyanide based on the injected chlorine in different contamination scenarios, various mathematical models were evaluated and the proposed equations for each model are presented in Table 2. With regards to the fitness indices of R2, RMSE and SSE, the Gaussian model provides the best functionality. In order to improve the prediction, the constant coefficients of the proposed Gaussian models (Table 2) were calibrated by single-purpose genetic algorithm based on Eq. 2 (minimizing the cost function). The calibrated model for prediction of residual cyanide in low contamination (2.5 mg L-1) scenario is presented in Eq. 4.

Table 2. Mathematical models for predicting residual cyanide based on the injected chlorine dose in different scenarios of contamination {(1) [CN-]=2.5 mg L-1, (2) [CN-]=5 mg L-1, (3) [CN-]=7.5 mg L-1}

Models	General Form	Senario	Typical Content	SSE	RMSE	R^2
		1	$a = 2.57$ $b = -0.3$ $c = -0.0002$ $d = 2.901$	0.01	0.05	0.97
Exponential	$RC^{*} = a \times exp\left(b \times CC^{\dagger}\right) + c \times exp\left(d \times CC\right)$	2	$a = 5.774$ $b = -0.365$ $c = 0$ $d = 0$	0.02	0.08	0.91
		3	$a = 840.1$ $b = -1.35$ $c = 0$ $d = 0$	0.01	0.03	0.95

Fourier	$RC = a_0 + a_1 \times cos(CC \times w) + b_1 \times sin(CC \times w)$	1	$a_0 = -1.357e+06$ $a_1 = 1.357e+06$ $b_1 = -583.5$ $w = 0.0004893$	0.01	0.07	0.96
		2	$a_0 = 2.851$ $a_1 = 2.273$ $b_1 = -1.059$ $w = 0.5098$	0.01	0.06	0.97
		3	$a_0 = 1.844e+05$ $a_1 = -1.844e+05$ $b_1 = -2447$ $w = 0.001915$	0.00	0.01	0.99
Gaussian	$RC = a_1 \times exp\left(-\left(\frac{(CC-b_1)}{c_1}\right)^2\right) + a_2 \times exp(-\left(\frac{(CC-b_2)}{c_2}\right)^2)$	1	$a_1 = 0$ $b_1 = -4.913$ $c_1 = 0.5982$ $a_2 = 2.481$ $b_2 = 0.1276$ $c_2 = 2.155$	0.01	0.01	0.93
		2	$a_1 = 5.113$ $b_1 = -0.3064$ $c_1 = 3.276$ $a_2 = 0$ $b_2 = 0$ $c_2 = 0$	0.97	0.01	0.97
		3	$a_1 = 2.292$ $b_1 = 3.877$ $c_1 = 1.307$ $a_2 = 0$ $b_2 = 0$ $c_2 = 0$	0.00	0.01	0.98
Rational	$RC = \frac{(p_1 \times CC^2 + p_2 \times CC + p_3)}{(q_1 \times CC^2 + q_2 \times CC + q_3)}$	1	$p_1 = -678.8$ $p_2 = -1193$ $p_3 = 1.037e+04$ $q_1 = 0$ $q_2 = 1$ $q_3 = 4179$	0.01	0.08	0.96

		2	$p_1 = 0$ $p_2 = -9.897$ $p_3 = 60.37$ $q_1 = 1$ $q_2 = -1.41$ $q_3 = 11.94$	0.01	0.04	0.97
		3	$p_1 = 0$ $p_2 = -2.532$ $p_3 = 15.72$ $q_1 = 0$ $q_2 = 1$ $q_3 = -2.161$	0.00	0.01	0.99
Polynomial	$RC = p_1 \times CC^2 + p_2 \times CC + p_3$	1	$p_1 = -0.1624$ $p_2 = -0.2855$ $p_3 = 2.48$	0.01	0.04	0.96
		2	$p_1 = 0.02926$ $p_2 = -1.2$ $p_3 = 5.357$	0.02	0.02	0.96
		3	$p_1 = 0.3383$ $p_2 = -4.688$ $p_3 = 16.08$	0.00	0.01	0.99

* RC = Residual cyanide concentration after injection of specified chlorine concentration (mg L-1)

† CC = Injected Chlorine concentration (mg L-1)

$$
\begin{gathered}
Gussian-distribution(For\ [CN^-]=2.5\,ppm) \\
[\textbf{Residual CN}] = \mathbf{a}_1 * \mathbf{exp}\left(-\left(\left([\textbf{NaOCl}]-\mathbf{b}_1\right)/\mathbf{c}_1\right)^2\right) + \mathbf{a}_2 * \mathbf{exp}\left(-\left(\left(\textbf{NaOCl}]-\mathbf{b}_2\right)/\mathbf{c}_2\right)^2\right) \\
-13.38 \le a_2 \le 18.34 \\
-15.76 \le b_2 \le 16.01 \\
-6.629 \le c_2 \le 10.94 \\
a_1 = 0 \\
b_1 = -4.913 \\
c_1 = 0.5982 \\
a_2 = 2.322 \\
b_2 = 0.3245 \\
c_2 = 0.9562
\end{gathered}
\tag{4}
$$

The abovementioned process has also been applied for cyanide contaminations of 5 and 7.5 mg L-1 (medium and high contamination scenarios). The calibrated models are expressed in Eq. 5 and Eq. 6, respectively.

$$Gussian-distribution(For\ [CN^{-}]=5\,ppm)$$
$$\left[\textbf{Residual CN}\right]\ =\mathbf{a}_1*\mathbf{exp}\left(-\left(\left(\left[\mathbf{NaOCl}\right]-\mathbf{b}_1\right)/\,\mathbf{c}_1\right)^2\right)$$
$$\begin{aligned}&4.174\le a_1\le 6.052\\&-1.498\le b_1\le 0.8852\\&2.268\le c_1\le 4.284\\&a_1\ =4.855\\&b_1\ =0.5691\\&c_1\ =4.023\end{aligned}\tag{5}$$

$$Gussian-distribution(For\ [CN^{-}]=7.5\,ppm)$$
$$\left[\textbf{Residual}\ \mathrm{CN}\right]\ =\mathbf{a}_1*\mathbf{exp}\left(-\left(\left(\left[\mathbf{NaOCl}\right]-\mathbf{b}_1\right)/\,\mathbf{c}_1\right)^2\right)$$
$$\begin{aligned}&0.3544\le a_1\le 4.229\\&2.444\le b_1\le 5.309\\&0.4596\le c_1\le 2.155\\&a_1\ =\ 2.075\\&b_1\ =\ 2.559\\&c_1\ =\ 1.694\end{aligned}\tag{6}$$

4. Conclusions

One of the main responsibilities of management systems and the provision of drinking water supplies, involve providing security against intentional or unintentional aspects which may threaten the quality and quantity of water. Cyanide may be found in the effluents of industries such as metal plating, synthetic fiber production and electronic manufacturers. This contaminant acts destructively upon human physiology, even in low concentrations (i.e. [CN-]>70 μg L-1), and can be used in sabotage operations. Cyanide's potential application for terroristic purposes due to its affordability and accessibility, besides the probability of its leakage from various industries increases the sensitivity upon this contaminant.

In the first step, the present study attempted to determine optimal concentrations of chlorine for the removal of cyanide contamination in water supplies with regards to three scenarios including low ([CN-]=2.5 mg L-1), medium ([CN-]=5 mg L-1), and high ([CN-]=7.5 mg L-1). The experiments resulted the optimal concentrations of chlorine as 2.9, 4.7, and 6.1 mg L-1 for low, medium and high contaminations, respectively. Moreover, the residual cyanide with regard to chlorine concentration for each of the contamination scenarios were mathematically modelled by applying various statistical distributions in the next step. Based on the obtained results, the Gaussian distribution was selected as the best model by evaluating the statistical measures of R2, SSE and RMSE. Finally, in order to calibrate the selected model, the square of difference between theoretical and experimental values were minimized by altering the coefficients of model, applying genetic algorithm.

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