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Research article

Chlorination of secondary treated wastewater with sodium hypochlorite (NaOCl): An effective single alternate to other disinfectants

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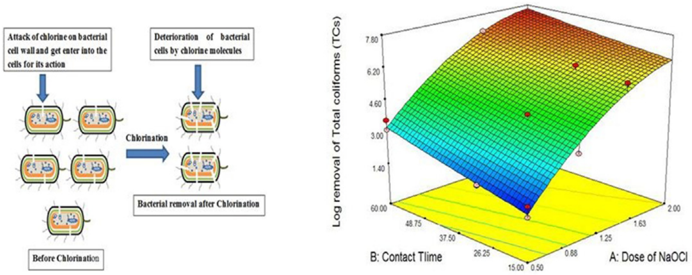
HIGHLIGHTS

- Efficient removal of *total coliforms* (*TCs*) with sodium hypochlorite (NaOCl) at 1.5 ppm concentrations during the time period of 15 min. with one-step chlorination.
- The chlorine residual maintenance under target value 1 mg/l.
- C_R* T concept studied in detail.
- The effect of pH on log removal rate of *TCs*, on rate of constant (k), and on dilution coefficient (n) was evaluated.
- Seasonal variations of MPN discussed.
- The Chick–Watson, Rennecker–Marinas, Collin–selleck, and Whites' modified kinetic modeling was applied to the secondary treated wastewater data for coliform removals, and to determine the effectiveness of the disinfectant.

GRAPHICAL ABSTRACT



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ABSTRACT

The present study provides details about the usefulness of chlorination in the recovery effluents of sewage, and to make it useable for irrigation purposes. Chlorination is one of the effective simplified, and cost-effective traditional methods for disinfection. The study was done for the period of March, 2019 to February, 2020. The disinfection process was optimized by adding sodium hypochlorite to the secondary treated effluents with the help of jar apparatus at a mixing speed of 100 rpm. To optimize the various process variables such as dose, and contact time, several concentrations of NaOCl (0.5, 1, 1.5, 2, 2.5, 3.0) ppm were carefully chosen at different time intervals of 15, 30, and 60 min respectively, which were centered on the foregoing studies. The factors like seasonal variation on MPN index of *total coliforms* (*TCs*), C_R * T concept, and effect of pH on log elimination of *TCs*, outcome of pH with rate constant (k), and results of pH against dilution coefficient (n) was also studied. The Chick–Watson, Rennecker–Marinas, Collin–Selleck, and modified Selleck models have shown good reliability to the experimental data of chlorine disinfection to be fit into these kinetic models for the treatment of sewage wastewater. The upgraded C_R * T values were attained by using disinfection models. Among these four models, the kinetic modeling by Collin–Selleck, and Selleck–White was investigated as the best modeling to be fitted more finely to the chlorination experimental data to count for the effectiveness of NaOCl. The selected indicator

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organism in the optimization process of chlorine was *Total coliforms* (*TCs*). The residual chlorine and most probable number per log unit (Log) for *TCs* were measured before the start and after the termination of the disinfection process. The World Health Organization (WHO) standard for pathogenic removal from wastewater, and to irrigate the crops is 3- to 4-log and the chlorine residual under 1 mg/l limit was accomplished.

1. Introduction

The heavy consumption of water has caused the water crisis with the increase in population, industrial load, urbanization etc. So, due to the high demands for water consumption, high rate of mortality, and health problems has been caused due to ill health of water distribution systems, and low hygienic conditions of the water distributing reservoirs (Yang and Zhang., 2013). Globally, around 884 lakh people were without the facility of fundamental need of potable supply of water in the year 2015 and about 2.3 billion populations was deprived off the vital cleanliness services such as of toilets or latrines (WHO, 2018a,b). The contamination of water can cause never-ending and severe ailments diarrhea, cholera, dysentery, typhoid fever, and polio among people (WHO, 2017). About 2 billion people get infected annually which is due to contamination of water and the annual death rate due to diarrhea only among children is around 502,000 (WHO, 2018a). The previous studies have shown that nearly 380 diseases were outreached due to the bloodsucking protozoans present in wastewater during 2011–2016 (Efstratiou et al., 2017). Furthermore, in South Asia and Sub-Saharan Africa the waterborne disease diarrhea is responsible for deaths around 87% (Troeger et al., 2017). Recently, in Aug, 2016, five thousand people out of fourteen thousand populations got illness due to campylobacter bacterium outbreak occurrence in ground water. According to U.S. Centers for Disease Control and Prevention (CDC, 2017) from year 2009–2014, the *Legionella* is recognized as the widespread microorganism, which is responsible for water causing diseases and deaths in U.S.A.

Therefore, wastewater treatment is a most considerable option to fulfill water demands and to protect mankind, and our environment. Hence, the sewage chlorination is an extensively used method of disinfection for treated wastewater. Chlorine in high amounts adversely affects human beings, if inhaled or exposed to it for long hours. The chlorine gas acts as a potential irritant and powerful electrophilic agent, which adversely affects the respiratory tract. It destroys the airways and distal lungs, and reasons for acute damages to respiratory tract. Its persistent nature is responsible for the respiratory ailment and lung inflammation (Hoyle and Svendsen, 2016). Chlorine based irritants straightly distresses the eyes and skin e.g. redness of eyes, tanning and allergy of skin, respiration problems etc. (Florentin et al., 2011; Li et al., 2015; Kanikowska et al., 2018). The products of chlorine leads to the growth of allergy related infections (Bernard, 2007). The exposure in long run to DBPs of chlorination at high concentration than the recommended dosage is also associated with certain cancers, liver, kidney, colon, and reproductive failures (Gopal et al., 2007; Hrudey, 2009; EPA, 2012). Although, chlorine leads to the formation of disinfection by-products (DBPs), yet it is a widely accepted way of disinfection, because, it is a simple, cheap, and an effective way of disinfecting wastewater (Hrudey, 2009; Watson et al., 2012; Ding et al., 2013; Liu and Zhang, 2014; Cai et al., 2016; Tak and Kumar, 2017) due to its great residual power to be retained in water/wastewater (McGuire, 2018), and it is also known as the strongest biocide, virucide, and anti-protozoan disinfectant (White, 2010). The *Legionella* outbreak can be restricted by chlorine and chloramines residual in water supply plumbing areas (CDC, 2017). The *E. coli*, a coliform bacterium is a well-known indicator microorganism for the assessing the quality of drinking water and reclaimed water reuse. The literature studies unveiled that the disinfection of wastewater with chlorine is quite effective by adopting one-step, two-step, and three-step chlorination techniques (Ding, 2010; Li et al., 2017a,b). The multi-step treatment of wastewater with chlorine may have improved the efficiencies of bacterial log removals just like 0.19-log, 0.20-log (Ding, 2010), 0.73-log (Li et al., 2017a), and 1.02-log (Li et al., 2017b) at a high dose of 6 ppm (Ding, 2010; Li et al., 2017a, b), which seems to be more complicated and prerequisites more attention. The process enhancements attained at very high doses that may lead to the generation of DBPs.

The literature revealed that reusable water is capable of having applicability in irrigation, bathroom flushing, landscape cultivation, clean-up uses, industrialized reuse, amusement works, recharging of groundwater table, and environmental enhancements (Asano et al., 1996; El-Gohary et al., 1998; Jiménez et al., 2001; Jimenez and Chavez, 2002; Scott et al., 2003; Chang et al., 2007; Yang and Abbaspour, 2007; Chiou, 2008; Huang et al., 2011). About 3- to 4-log removal of pathogens is a standard for irrigation practices to cultivate agricultural yields (WHO, 2006). The minimum EPA limit on chlorine residual for crop cultivation reuse is 1 mg/l (EPA, 2012).

The incorporation of C_R* T values concept is a strong fundamental research for the improvement of the disinfection process and as we have seen there is lack of literature too on chlorine optimization process with the help of C_R* T scenario, which might be a detailed topic of discussion in research. The research was mainly done with dose and time, but a few researchers had applied the C_R * T concept, which act as constant one value to fight with microbial populations under different environmental conditions and well offers the efficiency of disinfectant as well as microorganisms. Thus, we tried to discuss this concept in detail under this study.

Modeling of secondary wastewater chlorination is added in the manuscript, which had not been discussed considerably in literature findings. So, if we could apply these classic models to the treatment plants on regular basis, it may definitely help in improving the disinfection of wastewater. The fitting of data set to these classic disinfection models figure out in making of optimal designs for the complex disinfection processes and to progress the disinfection efficiency of the disinfection system (by increasing or decreasing the values of disinfection coefficients) and to control various complex phenomenon (e.g. Bacterial growth and decay, consumption of substrates, and hydrolysis of engulfed organic materials by bacteria) involved. These models were attempted to understand the interactions of disinfectant with the targeted molecular sites by microbes (Lyndon et al., 1999). Prediction of future of microbial inactivation is possible due to modeling of wastewater. Modeling helps in calibration and validation of various types of process kinetic parameters questioned for research, and aids in to find numerous inconsistencies in experimental as well as predicted parameters that may further help in planning for the seasonal circumstances. The kinetic parameters also act as coefficients database references (Serdarevic et al., 2016).

Henceforward, with the support of characterization of kinetics by developing a cost-effective disinfection system, and by effectively regulating a number of emerging DBPs of disinfection, we can positively recover the health of persons/animals as well as the well-being of environmental sustainability.

Accordingly, the accurately planned and proposed treatment of wastewater is desirable to mark wastewater as ecological for reusing purposes (Danial, 2002; EPA, 2012; Verma et al., 2015). Consequently, the high demand for water consumption, the human health concerns associated with contaminated water use, and the increasing issues related to aquatic life are some of the main considerable reasons to implant effective wastewater treatment. Therefore, disinfection is an indispensable mode to achieve higher log removal of microbes present in wastewater and to meet WHO guidelines for wastewater reuse.

The objective of the research is to find an economical, comprehensive way of secondary wastewater of sewage sterilization and to make it irrigable by using the least amount of chemicals with one-step chlorination. Also, the research aims to look into the C_R * T set-up for chlorination process in detail, and to validate numerous mathematical models for secondary treated wastewater chlorination. Therefore, the disinfection method for wastewater treatment with chlorine in this study is a matter of concern, because of its simplicity, great residual power, and keystone effects on microbial eliminations.

2. Materials and methods

2.1. Chemicals and reagents

The stock solution of sodium hypochlorite (NaOCl) was prepared by diluting the reagent grade solution of NaOCl purchased from Sigma-Aldrich having ~40,000 to 50,000 ppm active chlorine concentration. The stock and the working solutions of NaOCl were standardized with the help of an indicator solution of Di-ethyl-phenylenediamine (DPD) sulfate. The DPD sulfate was bought from Sigma-Aldrich. The absorbance was measured on the UV–Visible ranged Spectrophotometer (Systronics 119) at a wavelength of 515 nm with the help of a 1 cm capacity glass cuvette. The methods adopted for the characterization of influents and effluents were as recommended by (APHA et al., 2012), and were specified in Table 1.

2.2. Sampling location and collection of sample

The secondary treated wastewater was collected from the exit point of the plant, and the raw water was stored up from the inlet point of the Sewage Treatment Plant (STP) located on Ram Tirath road in Guru Nanak Dev University Campus, Amritsar, Punjab. The capacity of the sewage treatment plant was 2500 m^3/day. The secondary treatment of the STP was based on the aeration of wastewater by 15 hp aerators and sedimentation of the sludge in the secondary clarifier. The selection of twelve months study period for performing disinfection experiments was from March 2019 to February 2020. The samples of wastewater were collected during the peak times of 10–12 pm into the sterilized glass bottles and get transferred to the laboratory. Then, immediately, the samples were analyzed for disinfection study and the remaining samples were stored under dark conditions at about 4 °C to take further wastewater characterization experiments. The characteristics of raw and treated sewage water were mentioned in Table 2.

2.3. Experimental set-up

The disinfection experiments were conducted on secondary treated effluents of STP with the help of NaOCl disinfectant. The *Total coliforms* (*TCs*) were selected as indicator organisms for enumeration of disinfection efficiency of the disinfectant in use, because these indicate about fecal as well as non-fecal contaminants of wastewater (EPA, 2012). Based on prior studies and the initial experiments performed, the specified disinfectant amount range was selected between 0.5 to 3 mg/l. The 500 ml sample was added to the sterile glass beaker and the desired dose of NaOCl was added. The contents of the sample were mixed thoroughly for the complete dissolution of constituents of sample on jar apparatus at 100 rpm mixing speed of flocculator. The samples were withdrawn in sterile beakers after 15, 30, and 60 min intervals to analyze *Total coliform* counts and to conduct the experiments on free residual chlorine measurements. The initial free residual chlorine was checked after adding the required amount of NaOCl concentration and then followed by the MPN test technique differently to come across the MPN index. After the chosen period, the residual chlorine was checked immediately, and the MPN procedure was performed to test out the MPN Index of the desired sample. A total set of 9 disinfection experiments of MPN for *total coliforms* test as well as for residual chlorine, separately were conducted in triplicate for each dose at each interval of time for treatment.

Table 1. Methods adopted for different variables as per standards methods.

Applied parameter	Technique
pH	Equip-tronics Digital pH Meter Model EQ-610
Conductivity, µs/cm	2510 B. Laboratory Method with Labtronics Microprocessor COND-TDS-SAL Meter LT-51
Temperature, °C	Labtronics Microprocessor COND.-TDS-SAL Meter LT-51
Chemical Oxygen Demand (COD), mg/l	5220 C. Closed Reflux, Titrimetric
Biochemical Oxygen Demand (BOD), mg/l	5210 B. 5-Day BOD Test
Total Suspended Solids, TSS, mg/l	2540 D. Total Suspended Solids dried at 103–105 °C
Total coliforms (TCs), MPN Index/100 ml	9221 B. Standard *Total Coliforms* Fermentation Technique

[a] APHA et al. (2012).

Table 2. Characteristics of raw and treated secondary wastewater of sewage treatment plant (STP).

Parameter	Influent	Effluent
pH	7.65 ± 0.20	8.07 ± 0.25
Conductivity, µS/cm	980.50 ± 63.51	933.85 ± 93.32
Temperature, °C	26.49 ± 3.76	27.19 ± 2.89
Chemical Oxygen Demand (COD), mg/l	113.23 ± 68.96	48.32 ± 28.05
Biochemical Oxygen Demand (BOD), mg/l	34.84 ± 17.94	18.78 ± 10.59
Total Suspended Solids (TSS), mg/l	120.23 ± 52.12	30.49 ± 25.44
Total coliforms (*TCs*), MPN Index/100 ml	10^5–10^7	10^4–10^6

[a] Data values are represented as the mean standard deviation of the triplicate values.

2.4. The multiple fermentation tube method (MPN)

9221 B. Standard *Total coliform* Fermentation Technique was adopted (APHA et al., 2012). The quantification and qualification of *TCs* were evaluated during the study period and then applied to several kinetic models (Chick–Watson, Rennecker–Marinas, Collin–Selleck, and modified Selleck models). Lauryl Tryptose Broth medium (Himedia Company manufactured) was used. The media was freshly prepared to perform the MPN test. The serial dilutions (10, 1, 0.1) were used for maximum dose 3 ppm of NaOCl, and the highest dilutions (0.1, 0.01, 0.001, 0.0001, 0.00001, 0.000001, 0.0000001) were used in case of 0.5 ppm minimum applied dose of sodium hypochlorite. The dilution tubes were set in a row of five for each dilution in tube racks. The inverted Durham tubes were also added to culture tubes to check for any bacterial growth. The media, dilution water, and all other materials were autoclaved at 121 °C for 15 min. The sample and the respective dilutions were mixed well. The inoculation of culture tubes was done for each set of five culture tubes with replicate sample volumes differently in increasing order of serial dilutions and incubated at 35 ± 0.5 °C for 24 h. After, 24 h checked for the presence and absence of any growth, formation of gas, and occurrence of acidic reactions in the form of yellow pigmentation. The samples were further incubated for another 24 h means a total of 48 ± 03 h for the confirmation of positive presumptive test.

2.5. Action mechanism of chlorination and effect of pH on its functionality

Chlorine is well known for reduction of pathogenic microorganisms (bacteria, virus and protozoans etc.) present in sewage wastewater. Chlorine has been used in three forms i.e. gaseous chlorine, chloramines sodium hypochlorite, and calcium hypochlorite. Chlorine acts on the outermost layer of the bacteria and colipahges and broken up the complex chemical bonds made up of proteins and highly active enzymes. Among these four compounds sodium hypochlorite (NaOCl) is more common in use due to its stableness than the other ones. Its reaction take place with the addition of chlorine gas in water, where it gets quickly hydrolyzes to hydrochloric acid (HCl) and hypochlorous acid (HOCl) as expressed in equation (1), and equation (2).

$$Cl_2 + H_2O \leftrightarrow H^+ + Cl^- + HOCl \tag{1}$$

$$2\ NaOCl + H_2O \leftrightarrow HOCl + 2\ NaCl + H^+ \tag{2}$$

The pH plays a vital role in setting the ratio of hypochlorous acid to hypochlorite ion, and reduction of *coliforms* in chlorination of water and wastewater is explained in Figure 2a. The pH affects the rate of reaction of water (Cai et al., 2013). As we know that hypochlorous acid is a dominating ion at lower pH (less than or <7.5) and it has neutral charge on it and on the other hand, under higher neutrality conditions (>7.5 pH), the OCl^- ions are the major contributors in disinfection performance. Thus, during disinfection process, neutral charge of HOCl is more attacking on bacterial negatively charged surfaces and penetrates more easily into bacterial system. The active negative charge on OCl^- is responsible for low-slung disinfection, which is less reactive on pathogenic structures. The highly negative charged hypochlorite ions can minimize the exposure time of reaction (Connell, 1996; McGuire, 2018). Hence, by adjusting the pH of wastewater, the performance of the treatment plant can be increased and by increasing the susceptibility of bacteria and viruses to wastewater disinfection under lower pH or acidic conditions.

2.6. Kinetic modeling for chlorine

The Chick–Watson, Rennecker–Marinas, Collin–Selleck, and Selleck's kinetic model modified by White was handed down to find out the disinfection efficiency, and these models were compared with each other to examine their best fit with the experimental data.

2.6.1. Harriet Chick kinetic law improved by Herbert Watson (1908)

It defines that the product of concentration and contact time ($C_R * T$) for active and inactive microbes remains constant in disinfection process Equation (3). It denotes the negative slope for disinfection curve, which is proportional to $C_R^n * T$.

$$k = \lambda * C_R^n$$

$$dN_t/dt = -\lambda * C_R^n * N_t$$

Integrating the above equation, we get,

$$N_t/N_0 = -\lambda * C_R^n * T$$

$$\text{Log10 } N_t/N_0 = e^{-\lambda} C_R^n * T$$

This equation does not account for heterogenic variable wastewaters.

$$\boxed{\text{Log10 } N_t/N_0 = -\, k * C_R^n * T} \longrightarrow \textbf{Chick - Watson Equation} \tag{3}$$

k = inactivation constant for microorganisms, λ = die-off/microbial kill/specific lethality constant, C_R = final residual concentration of desired disinfectant, n = coefficient of the required dilution.If we got a rise in the temperature, then lower values of $C_R * T$ are required. And if we intensify the pH above 6, it converts the powerful HOCl acid to ClO^- ions. At higher pH, we lost HOCl and at lower pH the solution causes corrosiveness (Watson, 1908; Metcalf and Eddy, 2003).

2.6.2. Delayed Chick–Watson/Rennecker–Marinas kinetic law

This law is based on the inactivation of certain types of organisms with the increment of the $C_R * T$ values as mentioned in Equation (4). It is mainly applicable on oocysts, and endospores

$$\text{Log10 } N_t/N_0 = -\lambda\,(C_R * T - b)$$

$$\text{Log10 } N_t/N_0 = -\lambda\, C_R * T + \lambda\, b, \text{ (Slope} = -\lambda\text{, intercept } \lambda\, b)$$

where, b = lag coefficient mg * min/L; k= second order post shoulder inactivation constant.

$$\boxed{\text{Log10 } N_t/N_0 = -\, k * C_R^n * T + (k * b)} \longrightarrow \textbf{Rennecker - Marinas Equation} \tag{4}$$

Rennecker et al. (1999, 2001), Metcalf and Eddy (2003).

2.6.3. Collin–Selleck kinetic law (1972)

This chemical equation is used for disinfection of domestic waster for detection of TCs as well as for other bacterial species by new alternates of disinfectants. (Metcalf and Eddy, 2003). It is applied, where declining rate of disinfection exists as described in Equation (5)

$$\boxed{N_t/N_0 = (1 + 0.23\, C_R * T)^3} \longrightarrow \textbf{Collin - Selleck Equation} \tag{5}$$

where, N_t = number of microbes obtained after disinfection, N_0 = number of microbes before obtained disinfection, $C_R * T$ = chlorine residual at any time T, min (Collins et al., 1971, Collins and Selleck, 1972; Metcalf and Eddy, 2003).

2.6.4. Selleck kinetic Law (1978) modified by White (1999)

In this model, the lag and tailing effects are present in the combined form as defined in Equation (6).

$$N_t/N_0 = 1,\ \text{when, } C_R * T < b$$

$$N_t/N_0 \neq 1,\ \text{when, } C_R * T > b$$

$$\boxed{N_t/N_0 = [(C_R * T)/b]^{-n}} \longrightarrow \textbf{Selleck - White Equation} \tag{6}$$

where, $C_R * T$ = free residual chlorine remained at any time after the disinfection treatment, T = contact time, min, n = slope of inactivation curve (n value for secondary wastewater effluents for *TCs* = 2.8), b = x – intercept, when $N_t/N_0 = 1$, or $N_t/N_0 = 0$, and value of b = 4 for secondary wastewater effluents for *TCs* (Selleck et al., 1978; Metcalf and Eddy, 2003, White, 2010; Liang et al., 2013).

2.7. Historical findings

Table 3, provides the historic comparison of chlorination data with other disinfectants and it indicates that chlorination achieve maximum log reductions of microbes in comparison to other disinfection processes at low doses, because of its high residual powers than others.

On the other hand, Figure 1, highlights the historic dose optimization at some particular period with the help of the $C_R * T$ concept, as we know that the $C_R * T$ value provides the improved purification of wastewater. The $C_R * T$ values were calculated for historic data. Usually, this idea has applications in regulatory processes, but it could also apply to continuous wastewater cleansing activities. The historical range for $C_R * T$ data was between 0.042 and 685 mg * min/L. However, most of the researchers had suggested the best $C_R * T$ values under 100 mg * min/L. Our documented $C_R * T$ strength was 11.85 mg * min/L (1.5 ppm dose and at a contact time of 15 min). The high $C_R * T$ standards may enhance the process treatment, but its increment might also be a reason for the water/wastewater toxicity development problems. Consequently, the lower $C_R * T$ values were more preferable by considering other characteristic conditions of the wastewater.

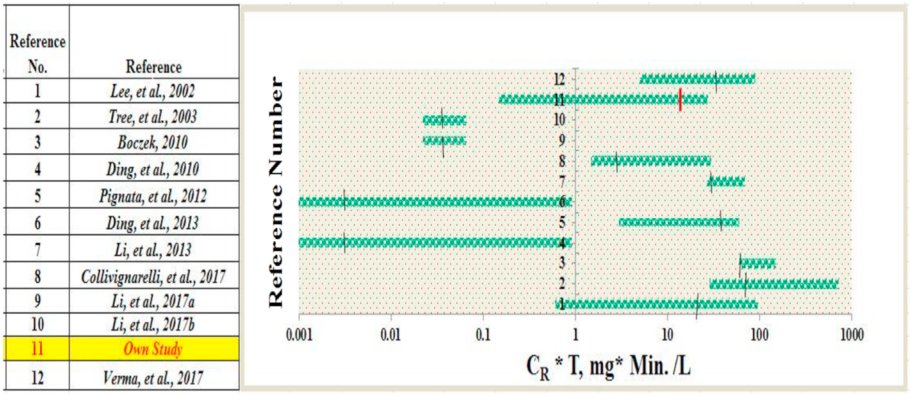
Table 3. Historical data collection on some of the major disinfection processes for wastewater treatment.

Sample Type	Dose, mg/l	Contact Time, Min.	Log removal rate, Log Units	Reference
Chlorination				
STP	8	5	>5	Tree et al. (2003)
Municipal Wastewater Treatment Plant (MWTP)	2	60	4.00	Blatchley et al. (2007)
MWTP	5	20	2.70	Lee et al. (2008)
STP Primary Saline Sewage Effluents	4.09	30	4.09	Li et al. (2017b)
Treated Urban Wastewater	2	60	>7.20	Rodríguez-Chueca et al. (2015)
STP (Secondary Effluents)	2.5	20	2.00	Verma et al. (2017), McGuire (2018)
Laboratory Culturable Sample	5	10	6.00	Xu et al. (2018), McGuire (2018)
Chlorine dioxide disinfection				
Waste Water Treatment Plant (WWTP)	1.5	25	>6	Sun et al. (2007)
Artificial Culture Sample	7	10	**3.74**	Vaid et al. (2010)
WWTP	3	30	**4.02**	Zhou et al. (2016)
Artificial Iceberg Lettuce Washing Water	3	0.5	**4.74**	Hassenberg et al. (2017), McGuire (2018)
Peracetic acid disinfection				
Municipal Wastewater Treatment Plant (MWTP) and Industrial Wastewater Mix	15	36	**4.00**	Mezzanotte et al. (2007)
MWTP	4	10	**2.00**	Beber de Souza et al. (2015)
MWTP	2.1	30	**3.00**	Bonetta et al. (2017)
WWTP	6	20	**4.50**	Garg et al. (2016)
WWTP	2	20	**1.80**	Garg et al. (2016)
Uv Disinfection				
WWTP	164 mWs/cm^2	15	**3.00**	Abou-Elela et al. (2012)
Treated Urban Wastewater	660 W/m^2	10	**7.50**	Rodríguez-Chueca et al. (2015), McGuire (2018)
Domestic Wastewater Treatment Plant (DWTP)	117 mWs/cm^2	10	**3.00**	McGuire (2018)
Water Treatment Plant (WTP)	89 mJ/cm^2	20	**2.40**	Garg et al. (2013), Garg et al. (2016)
Laboratory Culturable Sample	80 mJ/cm^2	30	**6.25**	Xu et al., (2018), McGuire (2018)
Domestic Wastewater	69.4 mJ/cm^2	412	**3.70**	McGuire (2018), Nguyen et al. (2019)
Ozonation				
MWTP and Industrial Wastewater Mix	5.3	6.4	**5.00**	Mezzanotte et al. (2007)
MWTP	20	15	**4.50**	Paraskeva et al. (1999), Paraskeva and Graham (2002), Bhatta et al. (2015)
Tertiary Wastewater Effluents	15	5	**5.74**	Lazarova et al. (2013)
WWTP	800	20	**8.91**	Bhatta et al. (2015)

3. Results and discussion

3.1. Effect of pH on reaction mechanism, rate constant, and dilution coefficient

Figure 2a demonstrates the log survival efficiency and Figure 2b determines the relationship for rate of reaction constant (k) in log subtraction of *TCs* upturns with the decline of pH. The rate constant also behaves well at high dilution coefficient (n) conditions as defined in Figure 2c. The rate constant for *TCs* survival rises with the advancement of dilution coefficient. The high k * C_R * T values improve disinfection (Li et al., 2017a). The high chlorine ions and decreasing pH of a reaction, counts the speed of a reaction in chlorination process. The pH has important role to control a reaction mechanism such as revealed in Figure 2a. The pH determines the type of water/wastewater whether the water is of acidic neutral or basic nature. The pH varies from hot weather



Reference No.	Reference
1	*Lee, et al., 2002*
2	*Tree, et al., 2003*
3	*Boczek, 2010*
4	*Ding, et al., 2010*
5	*Pignata, et al., 2012*
6	*Ding, et al., 2013*
7	*Li, et al., 2013*
8	*Collivignarelli, et al., 2017*
9	*Li, et al., 2017a*
10	*Li, et al., 2017b*
11	*Own Study*
12	*Verma, et al., 2017*

Figure 1. Literature findings for NaOCl as the product of concentration × contact time (C_R * T), mg * min/L (Boczek et al., 2010; Collivignarelli et al., 2017; Pignata et al., 2012).

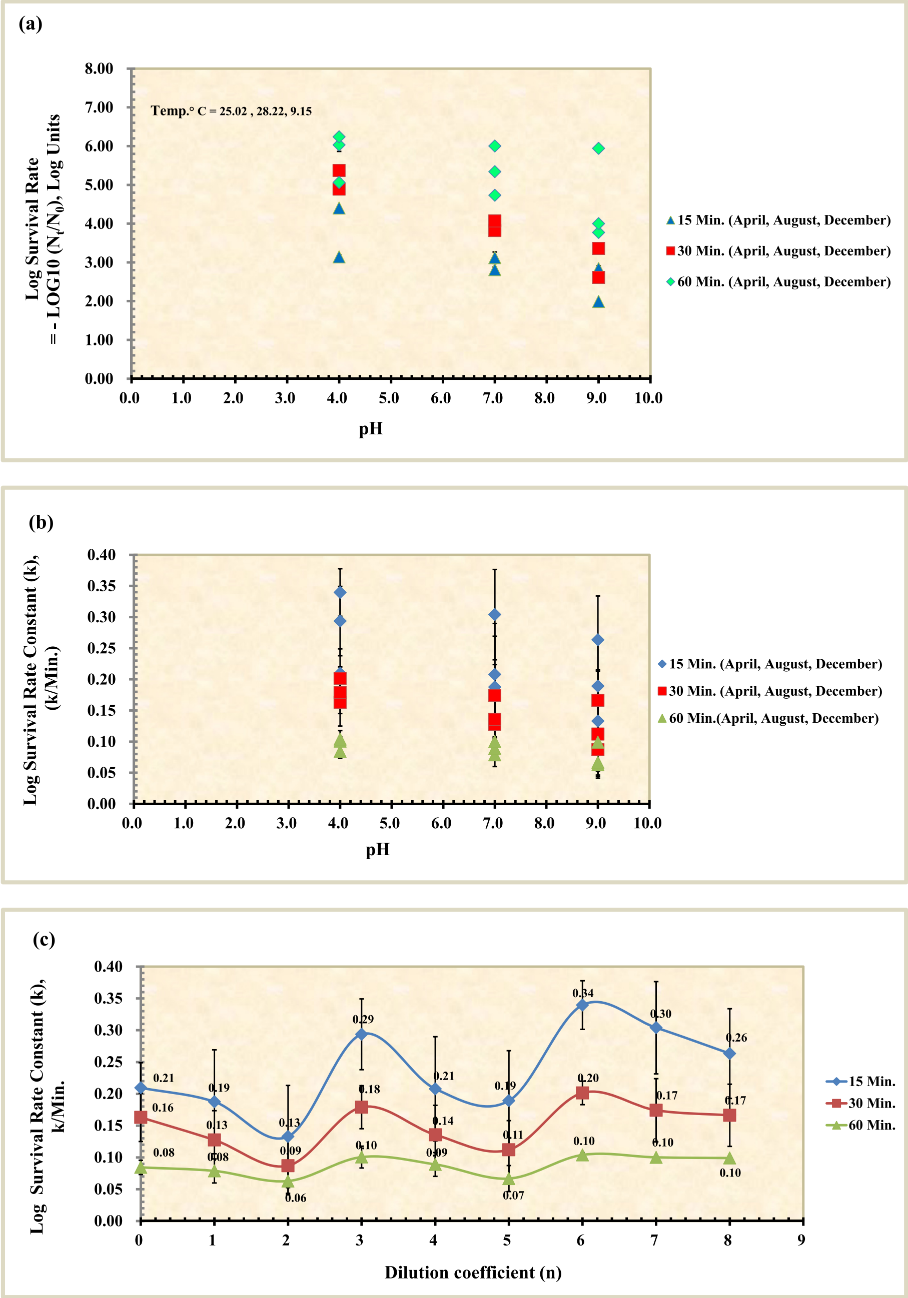


Figure 2. (a) Variations of pH w.r.t. log inactivation of *total coliforms* (*TCs*). (b) Effect of pH on reaction rate constant (k). (c) Influence of pH on dilution coefficient (n). Every outcome has happened at a dose conc. of 1.5 mg/l NaOCl, and over the exposure times of 15, 30, and 60 min during the months of April, August, and December, separately. Each error bars stipulates the standard deviation of triplicate monthly measurements.

to winter season. It decreases with the increase of temperature and increases with the decrease of temperature depending on the properties of water. As shown in Figure 2, the pH trend was grown from April (Hot month) to December (cold month), because pH varies from low to high during hot to cold seasons. The pH had gone up under acidic conditions followed by lowered down under alkaline conditions of wastewater. The highest chlorination disinfection was obtained underneath of acidic conditions (pH = 4) and the lowest disinfection was observed at alkaline (>7) atmosphere of wastewater. The concept of lower pH works better, but it had little effect on the STP under study, because the plant had maintained its neutral to alkaline conditions, and the pH varied not too much, and there was observed a good removal (>3-log) of coliforms at the lower set dose 1.5 ppm of NaOCl under these settings i.e. under acidic to neutral conditions. It was found, if any treatment plant could maintain its pH from neutral to alkaline, it may work better for disinfection process.

3.2. Disinfection efficiency in the form of total coliforms log removals

Figure 3a, b, and Table S1, validates the efficiency of the sodium hypochlorite disinfectant in counter to the elimination of *total coliforms (TCs),* which was improved with the developments of dose and time. The sewage samples were collected on different days over 5-months. Initially, before chlorine disinfection, the available population of *total coliforms* present into the treated STP effluent samples was ranged between the magnitudes of 10^4–10^6 MPN/100 ml. The graphical representation shows the consistent rise of the efficacy of the disinfectant during the disinfection optimization study experiments with the enhancement of the

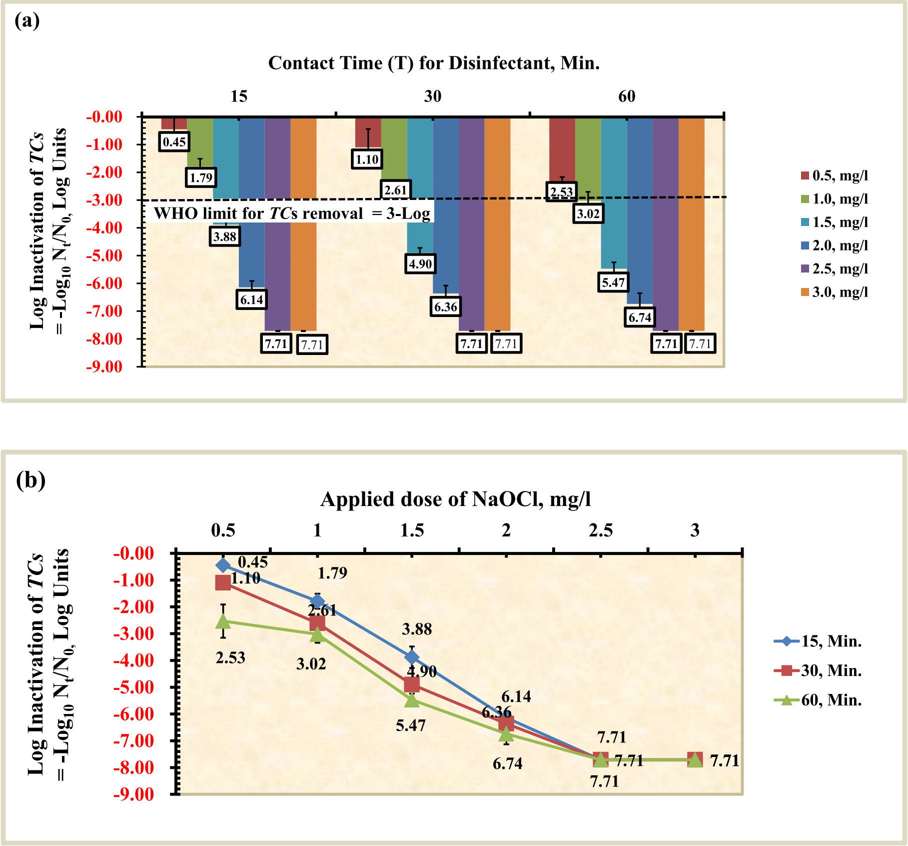


Figure 3. (a) Log deactivation of *Total coliforms* (*TCs*) from secondary treated wastewater of STP at different intervals of time, (15, 30, and 60) minutes. (b) Log inactivation of *TCs* at diversified dose applications of disinfectant i.e. 0.5, 1, 1.5, 2, 2.5, and 3 mg/l. Square dotted dash line state the WHO limit for agricultural wastewater re-uses. Each error bars specify the standard deviation of triplicate dimensions.

dose and time. The lowest log reduction perceived was 0.45-log with a dose of 0.5 mg/l at a contact time of 15 min, and the highest log reduction of 7.71-log was given away at a dose of 2.5 ppm as well as for 3 ppm over 15 min time period, respectively. The 1.5 ppm NaOCl dose was considered as the optimized dose, which has to get hold of the targeted norms (1000 MPN/100 ml or 3-log removal) over the exposure times of 15, 30, 60 min, individually. The MPN index 3.88-, 4.90-, 5.47-log was attained at the adapted dose, 1.5 ppm during the treatment over 15, 30, and 60 min, similarly. There were no *TCs* progression or prolonged disinfection brings into being at 2.5 ppm measured quantity after 15 min treatment time and more than 7.71-log elimination of bacteria was obtained. It was seen that the *TCs* found to be sensitive to chlorine species. Almost similar type of trends was achieved for all the preferred dosages. The nominal log reduction was observed may be due to the presence of chlorine demand for oxidizing matter into the sewage samples. The rapid elimination of bacteria was happened within first 15 min that may be due to the quick action of HOCl, an active chlorine species that works rapidly on oxidizing compounds and bacterial population. The growth of bacteria acted asymptotically after 15 min at 2.5 ppm concentration, which specifies no happening of variations among bacterial inhabitants of wastewater. The overall or 100 percent or >7-log removal of *TCs* was reached at dose application of 2.5 mg/l after 15 min. The similar trends of results for decay of *TCs* were also attained after 2.5 ppm or higher concentration of disinfectant like 3 ppm, which means the bacterial evolution, undergoes to tailing/log effect, which indicates complete kill of microbial populations, and there was no happening of activation of *TCs*. This type of tendency was found in every concentration beyond 2.5 ppm dosage of NaOCl, and this style of bacterial proliferation was also driven up with the time.

The historical literature articulated that a 2–6 ppm dose of chlorine was the best option to get reclaimable water (Li et al., 2017a,b; Verma et al., 2017; Xu et al., 2018; McGuire, 2018). A research advised that the substantial removal of *E. coli* (6.15-log) at 2 mg/l over 15 min and the complete removal >6-log (0 < 1.8 CFU/100 ml) was accomplished during the 60 min treatment (Rodríguez-Chueca et al., 2015). One more study had proposed that the deductions of *E. coli* at a minimum dose of 4 ppm at 30 min, and a maximum dose of 6 ppm at 15 min, consistently. (Li et al., 2017a,b). In an another significant work, for bringing down the *TCs* genera, the cure was performed on STP samples with a high chlorine dose of 4 ppm and a contact time of 20 min (Verma et al., 2017; McGuire, 2018). According to a bench-scale disinfection, the 6-log decline of *E. coli* was examined by applying a dose of 5 ppm at 30 min (Xu et al., 2018).

Our proposed dose was 1.5 ppm at exposure time of 15 min, because it complied with the anticipated goal line at this prescribed amount. In this manner, by lowering the dose extent, and lengthening the time period, a better disinfection was discovered.

3.3. *Usefulness of NaOCl as a role of residuals at any time*

Figure 4a, and Table S1, illustrates the effectiveness of NaOCl for killing germs as a function of free residual chlorine with time, which indicated that the chlorine residual lowers down with the progression of time. It was suggested that the 15 min residual was very crucial to estimate the efficiency of the disinfectant as given in Figure 4a. The prescribed standard for chlorine residual was obtained at 2 ppm NaOCl during 15 mintreatment, but the improved chlorine residuals were seen with the initial conc. of chlorine at 1.5 ppm over the time lapse of 15 min, which was under the set criteria (0.5–1 mg/l) of EPA/WHO. Figure 4b, explains

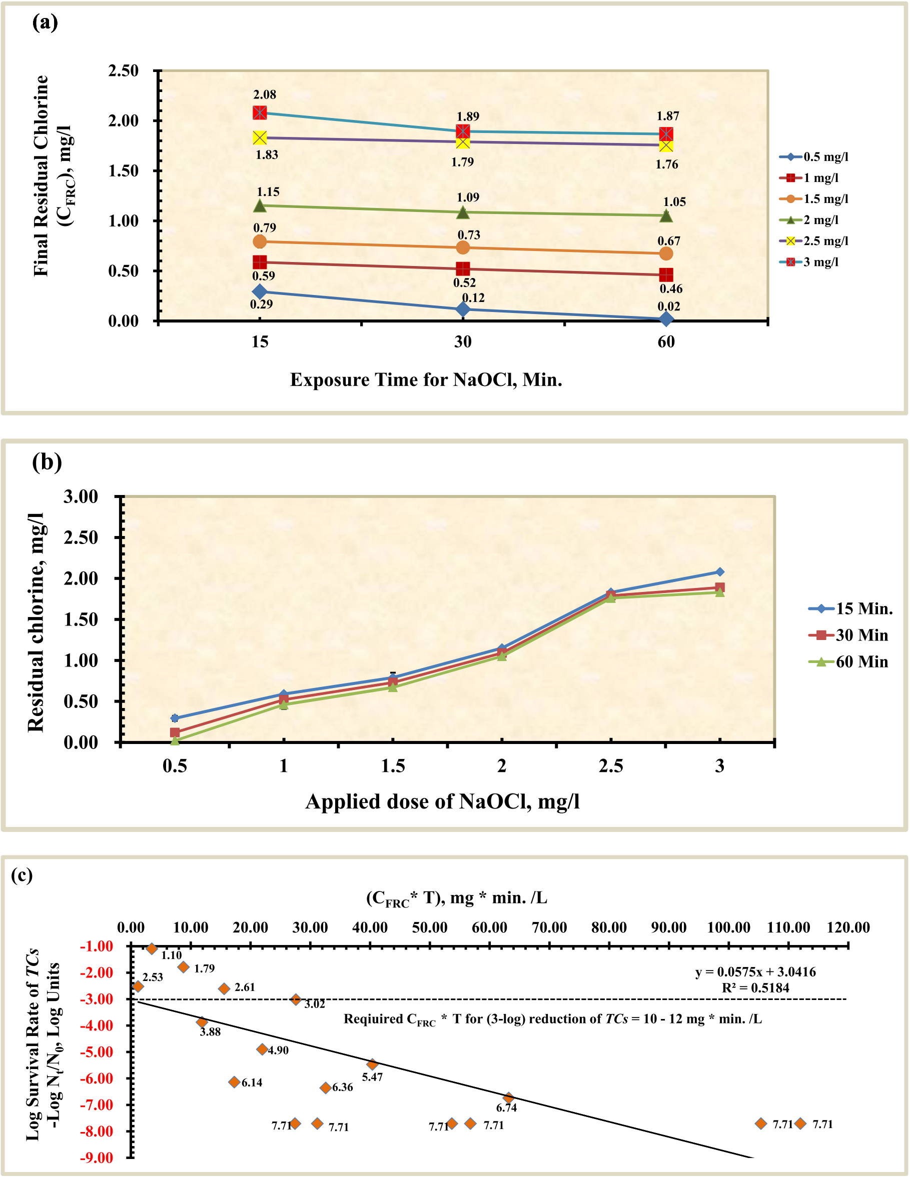


Figure 4. (a) Variations of residual chlorine with time. (b) Breakpoint chlorination curve of chlorine. (c) Efficacy of NaOCl as a function of C_R * T approach. Square dotted dash line postulates the target value for agricultural wastewater re-use. Each error bars signifies the standard deviation of triplicate values.

the chlorine residual with the practiced dose concentrations like 0.5, 1, 1.5, 2, 2.5, 3 ppm. The trend progressed up with the up-rise of applied dose of disinfectant. This curve is also known as chlorine breakpoint chlorination curve, because at the breakpoint, great amount of chloramines get oxidized and intensification of chlorine residuals occurred by continuous additions of chlorine, which further rises the residuals available as free chlorine. After the breakpoint occurrence, the remained chlorine residuals behave as the free chlorine residuals. The breakpoint was gotten at 0.79 mg/l, where the initial conc. of NaOCl as Cl_2 was 1.5 mg/l. The C_R* T conception further suggests that the concentration and time were equally predominant for achieving the usefulness of the disinfectant (Figure 4c.). The germ-killing efficiency was enhanced with the rise of the C_R* T level of the disinfectant. Nearly, all the concentration-time data has shown some of the little lag outcome, after that certain linearity in trends was experienced. The decline in concentration caused the lag results. Henceforth, the specified applications of dosage must be wanted to complete the deactivation of bacteria. The lag/shoulder, log/tailing, and combined shoulder-tailing effects were perceived for the disinfectant behavior on the bacterial populace. The lag/shoulder and log/tailing effects arose with the upgrading C_R* T standards. During the lag period, chlorine reacted with the suspended constituents of the wastewater. The bacteria undergone slow constant exponential growth and tried to adapt environmental conditions. They grew-up much in size, but not in numbers, so, there was no reduction in the microscopic populace. Dilution of samples helps in the growth of bacteria. The cells matured logarithmically throughout the log stage due to the accessibility of more nutrients for their development. In the ending phase of life or the stationary or at death point of microbial life, shielding of microorganisms befallen due to the presence of large particles and there happened no growth of cells due to slow metabolic activities of injured cells. The cells remained in inactivated form or they got death in water/wastewater. This is called the tailing effect of bacterial growth. This may take place due to the disinfectant attack or some other environmental factors.

The required C_R * T was ~10 to 12 mg * min/L to accomplish 3-log reduction of bacteria. The optimum effectiveness of sodium hypochlorite obtained was 3.88-log at a minimum C_R * T value of 11.85 mg * min/L, which means that the average residual chlorine calculated was 0.79 mg * min/L over a minimum time of 15 min.

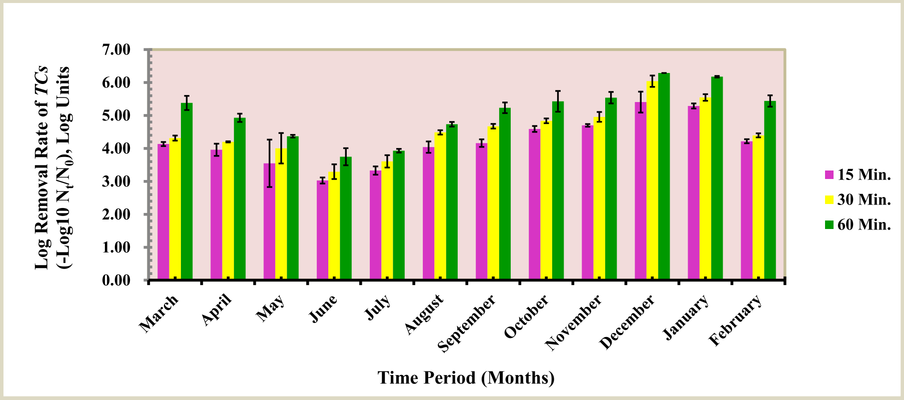


Figure 5. Seasonal variations of log removal rate of *total coliforms* (*TCs*) w.r.t. different time interval (15, 30, 60 Minutes). Each error bars states the standard deviation of triplicate monthly dimensions, individually.

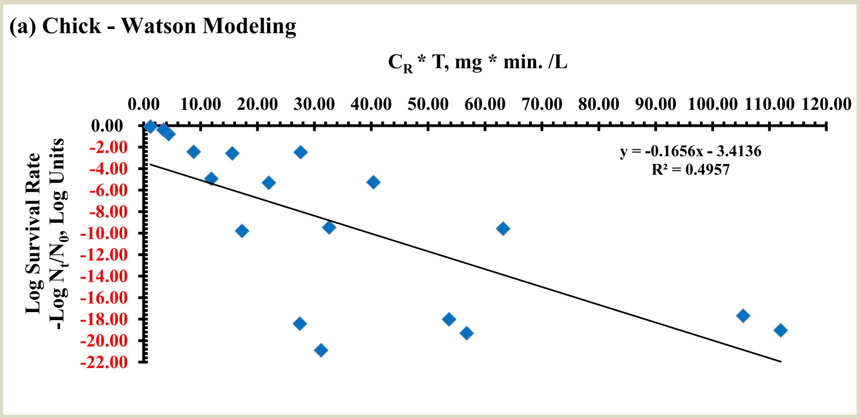
Previously, the literature has investigated that the 3-log removal of *TCs* was reduced at a $C_R * T$ value of 15 mg * min/L or log $C_R * T$ of 1.18 mg * min/L (Li et al., 2002). Another research examined that up to 3-log deduction of *TCs* was able at $C_R * T$ of 27.6 mg * min/L or Log $C_R * T$ 1.44 mg * min/L from reclaimable water of Municipal Treatment Plant (MTP) (Li et al., 2013). We discovered more than 3-log (i.e. 3.88-log) of *TCs* at 11.85 mg * min/L or Log C_RT 1.08 mg * min/L. Our results for $C_R * T$ values were consistent with the historic findings of literature (Li et al., 2002).

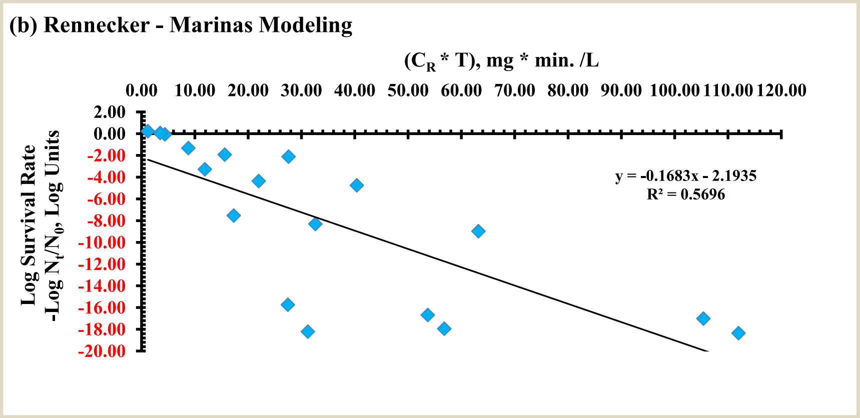
3.4. Seasonal effect of log removal rate of Total coliforms on chlorination disinfection

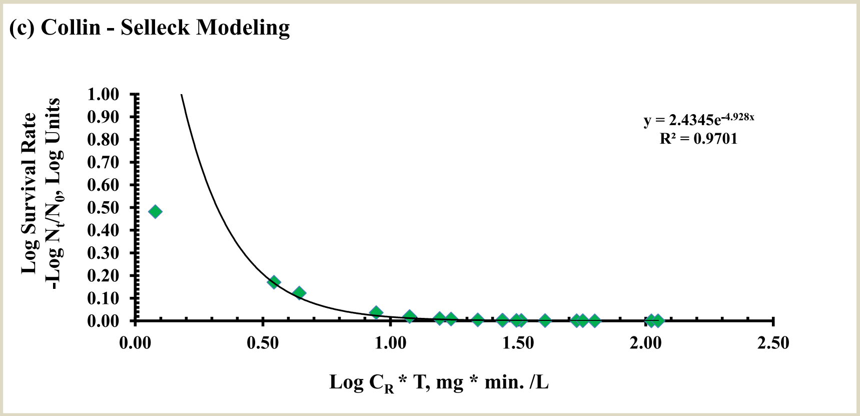
Figure 5, depicts that the chlorination disinfection efficiency leveled up during the cold months/medium cold i.e. from December to April due to cool conditions of weather and leveled down in the hot weather/hot humid weather from June to November due to hot and humid weather. The best log inactivation of *coliforms* was observed during the months of December and January followed up by February, March, April, and May, which was credited to the lower progression of bacteria during the wintertime and the highest deactivation (5.41-, 6.04-, 6.29-log for the contact time of 15, 30, and 60 Minutes) of *TCs* was recorded during the month of December at the time intervals of 15, 30, and 60 Minutes). Whilst, the lower log elimination of bacteria was verified during the months of June and July following August, September, October, and November, which was attributed to the great multiplication of bacteria during warm season. The lowest log reduction (3.03-, 3.29-, 3.75-log) for *TCs* was noted down in the month of June. As we know, the hot temperatures are usually works great for bacterial growth, which had dropped down the rate of inactivation of microorganisms. The 30 Minexposure time had provided the best treatment at all. Although, 60 mininterval had reached the great removals of *TCs*, yet it seems to be a long exposure time for disinfection process, because a disinfectant could works well during the initial 15–30 min for microorganisms. The seasonal variations are consistent with the study documented in literature (Lee et al., 2008).

3.5. Kinetic models and their validation

The kinetic modeling of secondary wastewater of sewage with chlorine states us about the action of mechanism of chlorine with constituents of wastewater and available free residual chlorine exist in it for continuation of the disinfection process. The data related to kinetic modeling represented in Table S1. In Figure 6a, the Chick–Watson kinematics represents the exponential growth of microorganisms or the early lag effect, which means initially, there was very low or no disinfection occurred due to the characteristics of wastewater microbes, and the applied disinfectant as descried in Equation (3). The poor mixing, delayed interactions of disinfectant with microbes on the targeted sites may be some of the factors responsible for lag effect occurrence in inactivation of *TCs*. After this lag effect, the linearity in the disinfection was happened with the improvement of $k * C_R * T$ values. As presented in Table S1, the k value drops with time and rises with strengthening of dose, and in response to it, the log inactivation of *TCs* was also got up. As displayed in Figure 6b, the straight line fit curve was obtained showing pseudo-first order kinetics or bimolecular reactions having order one with pseudo-first order rate constant (λ), which specifies that the overall rate of reaction is of second order; this was due to the presence of excessive amount of one of the reactants (Rennecker et al., 1999). A kinetic analysis was carried out on *B. paratyphosus* disinfection with phenol disinfectant (Chick, 1908). The initial lag effect (means no disinfection state) was stunned by the functioning of lag coefficient (b) in Rennecker–Marinas locomotive modeling. The Rennecker–Marinas motive balance further followed the Chick–Watson motile modeling as for the value of b approaches to zero. The Rennecker–Marinas and Chick–Watson kinetic equivalences were related to each other in the form of the lag coefficient as stated in Equation (4). It was explored in a study that when, $b = 1, 2, 3\ldots$so on, the delayed Chick–Watson mobile association works, but if $b = 0$, then the delayed Chick–Watson kinetic equivalence get converted to Chick–Watson (Wahman et al., 2009). The value used for dilution coefficient $(n) = 1$ in delayed Chick–Watson kinetics, which indicates that the fixed value of dilution coefficient (n) leads to the fixed inactivation of microbes by providing fixed product values of $C_R * T$. The value of dilution coefficient equals to 1, points out that the situation underrates bacterial inactivation, and when the value of dilution coefficient was not equals to 1, it overvalues the bacterial reductions (AWWA, 1999; Lee and Nam, 2002). The lag coefficient (b) discussed in delayed Chick–Watson mathematical problem had accounted for the consistency of the product of $C_R * T$ during disinfection treatment of wastewater or to overcome the lag/shoulder effects for the process of disinfection (Wahman et al., 2009). Our results were also consistent with Wahman et al., 2009. We have used Rennecker–Marinas energy moving modeling for halting of *total coliforms* with the aid of sodium hypochlorite disinfectant to accelerate chlorination kinetics on secondary treated wastewater of STP and it was noticed that both the lethality coefficient (λ), the lag coefficient (b), and the dilution coefficient (n) have played the important roles for up regulation of *total coliforms,* and in the process augmentation by overcoming the lag effects. On the other hand, Figure 6c, clarifies that the Collin–Selleck model elaborates the tailing effect (means reduced or no growth of bacteria) more noticeably, but it get lacked in enlightening the lag/shoulder effect, meanwhile, the combined lag and tailing effects of bacterial growth were effectively recognized through Selleck–White kinetic modeling for disinfection of secondary wastewater as provides in Figure 6d. The learning *about Total coliforms* deactivation has investigated the kinetics of chlorine disinfection (Selleck et al., 1978). The Selleck–White empirical model is applicable to different types of


(a) Chick - Watson Modeling


(b) Rennecker - Marinas Modeling


(c) Collin - Selleck Modeling

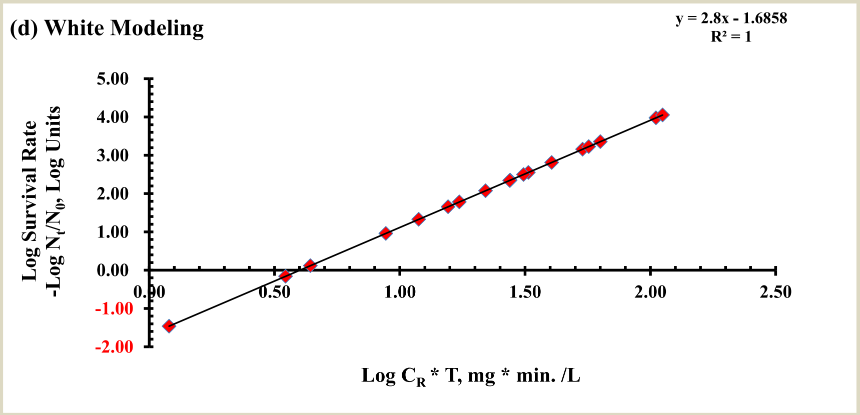

(d) White Modeling

Figure 6. (a, b, c, d) Fitting of experimental data into various types of classic kinetic models, where, n = 1 used for Chick–Watson, and Rennceker–Marinas kinetic models, and value of n = 2.8 and b = 4 used for Collin–Selleck, and refined Selleck or Whites' dynamic models. Value of inactivation constant k is given in Table S1.

disinfectants, entities in wastewater, and the testing wastewater samples (Lee and Nam, 2002). When, the C_R * T value rises to 100 mg * min/L, then the Collin–Selleck equalization i.e. Equation (5), behaves alike Selleck–Whites' mathematical statement viz. Equation (6), (Metcalf and Eddy, 2003). With the increment in intercept − b in modified Selleck kinetics works as displayed in Equation (6), the disinfection effectiveness could also be boosted-up. The upsurge in the values of dilution coefficient (n) in Collin–Selleck, and White kinematic models leads to the improvement of disinfection process as compared to the Chick–Watson and Rennecker–Marinas kinetic balancing. The refined Selleck kinetic modeling was discovered to be the most effective dynamical modeling sum for chlorine disinfection amongst the four preferred disinfection simulations, because it effectively counts both of the lag and log effects of microbial growth and decline Equation (6). It was shown as an excellent statistically ($R^2 = 1$) correlated energy model. Hence, the greater values of k * C_R * T, along with lag coefficient (b), dilution coefficient (n) and lethality coefficient (λ) helps in rising the prevention usefulness against micro flora. The dynamic models of Chick–Watson and Rennecker–Marinas were considered the best statistically fitted representations for chlorine disinfection, for the reason that they are more correlated with wastewater constituents. The enhanced k * C_R * T values, in addition with some other parameters such as lethality coefficient (λ), in case of Chick–Watson, and delayed Chick–Watson dynamics, the lag coefficient (b) for delayed Chick–Watson, and Whites' equalities, and dilution coefficient (n) for almost all the four forceful modeling evaluations were some of the determining factors in up-gradation of the rate of reaction in any disinfection process, which also further helped in calculating the better-quality measured quantities of disinfectant, and comparing the varied disinfectants one with each other. Although, the Chick–Watson and Rennecker–Marinas energetic paradigms were verified to be the best statistically fit actual energy models as they were known for lag effect removals, where k value has played the major role in estimating the microbial dismissal in driving equations, but in case of delayed Chick–Watson pattern, the assessment of n was also the noteworthy aspect. Yet, Collin–Selleck model distinguished the tailing effects in added prominent ways. Whites' potential energy model has given away great overexpression curve for *total coliforms* reduction in our experimental data, because of the reason that it significantly counts for the lag and tailing effects during the process of disinfection. The efficiency of one-phase chlorination was attributed to the good quality of wastewater, better mixing conditions under neutral to alkaline environments, and the lowest sensing of accessible free chlorine for taste-producing organic pollutant load into the sewage effluent water. As we know that the lower organic matter present in the sewage wastes always leads to the better disinfection of water/wastewater, as it has a low threshold for taste, which provides enhanced k * C_R* T values, pronounced synergistic effect (collective act of free chlorine and chloramines), and shortest regaining phases to microbes. The results demonstrated that the one-step chlorination was proved to achieve the disinfection goal and recycling of secondary treated wastewater of STP, which can be used further in irrigation of crops and other environmental developments.

4. Conclusions

The disinfection goal for wastewater recycling and agricultural reuse was accomplished with one-step chlorination at an optimized C_R* T value of 11.85 mg * min/L. The C_R * T concept offered a constant one value to fight with microbial populations under different environmental conditions, and good efficiency for disinfectant as well as microorganisms. The addition of k into the normal C_R * T values has refined up the C_R* T conception.

All the four classic kinetic models ascribed as one of the elementary models of disinfection were well fitted into the experimental data of secondary treated wastewater of sewage and the white' model was the best amongst all. The disinfection could be improved by making changes in the values of disinfection coefficients. The n, and b coefficients were noted to be stronger than k. The calculations, which have larger b, and n standards acted remarkably in chlorination model developments.

Declarations

Author contribution statement

Sunita Kesar: Conceived and designed the experiments; Performed the experiments; Analyzed and interpretted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Manpreet S. Bhatti: Conceived and designed the experiments; Contributed reagents, materials, analysis tools or data.

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Data availability statement

Data will be made available on request.

Declaration of interest's statement

The authors declare no conflict of interest.

Additional information

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