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## RESEARCH ARTICLE

### Mathematical Models for Chlorine Decay in Bulk Water Reservoirs

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#### Abstract

A brief idea of almost all models available in the literature for free chlorine concentration in water reservoir with first order decay rate and  $n^{\text{th}}$  order decay rate constants is given. A multiple reaction constituent model for the chlorine with fast reacting agents and slow reacting agents is presented and their effect on free chlorine is discussed. To see the effect of diffusion in  $x$  and  $y$  direction together with chlorine decay rate, two model are considered and the solutions for these models obtained analytically and their graphical representation and numerical values for different values of time distance, diffusivity and chlorine decay rate constant are obtained using MATLAB.

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#### Introduction

All people whatever their stage of development and their social and economical conditions, have the right to have access to an adequate supply of safe drinking water. One of the major World Health Organization (WHO) function to achieve such goals is the responsibility to propose regulations to make recommendations with respect to international health matters. The most popular disinfectant is chlorine which is used to kill pathogenic bacteria during to treatment of drinking water process all over the world. Chlorine also plays an important role in the maintenance of water quality in the distribution system through a residual. The distribution system consist of two components, one the water storage tank or reservoir and second the pipe line system. These two components differ significantly from each other as the water retention times and water flow properties are different in both these systems.

There are a number of research paper published on chlorine concentration decay in drinking water distribution system. Clark et al. (1994) showed how chlorine residuals can vary throughout the day at different locations in the distributive systems. Clark et al. (1995) used first order kinetics and rate of chlorine decay in their model. They showed that the fluid velocity and pipe radius affect the propagation of chlorine residual levels, disinfection efficiency and the formation of disinfection by-products. Reddy et al. (1996) discussed the weighted least-square method for some parameter estimation in water distribution network, like model pressure heads, pipe flow, head loss in pipes and consumptions in flows. David and Bryan (1996) developed an adjective transport model by neglecting the contribution of radials as well as axial diffusion terms. Munavali and Mohan (2005) presented a simulation-optimization model for water quality parameter estimation in the distribution system under dynamic state. Osman, and Metin (1999) solved two dimensional convection dispersive equation numerically for various boundary and initial conditions, considering the decay of chlorine in the bulk flow, but they did not consider the transfer of chlorine from bulk flow to the pipe wall. Jaipal et al. considered one and two dimensional steady state and unsteady state mathematical models that incorporate chlorine decay within the bulk flow and transport of chlorine decay within the bulk flow to the pipe wall and predicted the chlorine concentration in the drinking water distribution system. Chemical decay kinetics is concerned with the rate of reactions. Many reactions have rates, which at a given temperature are proportional to the concentration of one, two or more of the reactants raised to a small integral power (Swayer, et al., 1978). Chlorine disappears due to its reactions with ammonia and organic compounds naturally present in source water. In the water distribution system chlorine also reacts with the pipe wall (Viljeon et al., 1997; Hua et al., 1999). Various decay model exist, but the most pronounced used in chlorine decay is the first,

second and nth-order chlorine decay models. Boccelli et al.(2003) presented that chlorine disinfectant addition must achieve an adequate inactivation of pathogens before the treated water reaches the first customer(primary disinfection) and be large enough to ensure an adequate residual at the periphery of the distribution system to inhibit bacterial regrowth( secondary disinfection). As a result, and according to water quality regulations, it is essential to have minimum chlorine residual over the whole distribution system at all times. However, depending on the quality of water, types of treatment process and condition of distribution system chlorine decay behavior is significantly variable. Thus the chlorine demand the reaction time and its required set point or initial dosing vary from one water source to another and also for different water network. Chang et al.(2006) suggested that the reaction between chlorine and natural organic matter (NOM) leads to the formation of disinfection by-products (DBPS), some of which have been identified as potentially carcinogenic, mutagenic or toxic substances.

### Backgrounds of Chlorine Decay Models

Many research paper published on chlorine concentration decay in drinking water distribution systems are focused on the simplicity of the applications. Most water distribution network modelling packages assumed that chlorine decays by first order kinetics. The available non empirical chlorine decay models in the literature can be review as follows.

#### Single Constituent Chlorine Decay Models

Chlorine decay by first order kinetics with respect to chlorine can be defined by following equation

$$\frac{dc}{dt} = -Kc \quad (1)$$

Initial condition

$$c = c_0 \quad \text{at } t = 0 \quad (2)$$

Where  $c$  is chlorine concentration at time  $t$  (mg/l) and  $K$  is first order chlorine decay constant(per hour) .

The solution of above model is

$$c = c_0 e^{-Kt} \quad (3)$$

#### $n^{\text{th}}$ Order With Respect To Chlorine

$n^{\text{th}}$  order with respect to chlorine can be given by the equation

$$\frac{dc}{dt} = K_n c^n \quad (4)$$

Initial condition

$$c = c_0 \quad \text{at } t = 0 \quad (5)$$

Where  $K_n$  is  $n^{\text{th}}$  order chlorine decay

The solution for this model is

$$c = \frac{1}{\left[ K_n t (n-1) + \left( \frac{1}{c_0} \right)^{n-1} \right]^{\frac{1}{n-1}}} \quad (6)$$

If  $n=2$  (for second order chlorine decay model) then

$$c = \frac{c_0}{1 + c_0 K_2 t} \quad (7)$$

#### Two Constituent Chlorine Decay Models

Clark(1998) a two component second order chlorine decay model considering the effect of reactant constituent. Assuming the reaction between chlorine and other reacting substance. In short this model can be given as



$HOCl$  Generally reacts with various components that make up chlorine demand as follows



In equation (9), if the balanced reaction is represented by



Where A and B are reacting substances; and P is product of the reaction, then the rate of reaction is given by

$$\frac{dC_A}{dt} = -K_A C_A C_B, \quad \frac{dC_B}{dt} = -K_B C_A C_B, \quad \frac{dC_P}{dt} = K_P C_A C_B \quad (11)$$

$$\text{Where } \frac{K_A}{a} = \frac{K_B}{b} = \frac{K_P}{P}$$

If  $C_{A0}$  and  $C_{B0}$  represent the initial concentration of A and B, respectively, at  $t=0$ , and  $x$  represents concentration

of A that has reacted, then the concentration of B that has reacted is given by  $\frac{bx}{a}$ , and

$$C_A = C_{A0} - x, \quad C_B = C_{B0} - \frac{bx}{a} \quad (12)$$

$$dC_A = -dx = \frac{b}{a} dC_B \quad (13)$$

The solution of these equations is

$$C_A = \frac{K}{1 - Re^{-\alpha t}} \quad (14)$$

Where  $K = C_{A0} - \frac{a}{b} C_{B0}$  ( $mg / lt$ ),  $R = \frac{a C_{B0}}{b C_{A0}}$  dimensionless parameter,  $\alpha = \left( \frac{b C_{A0}}{a} - C_{B0} \right) K_A$  (per unit

time)

If  $c$  is the concentration of free chlorine then

$$c = \frac{K}{1 - Re^{-\alpha t}} \quad (15)$$

Where  $c$  is concentration of free chlorine in water in  $mg/lt$  at time  $t$ .

### Two constituent chlorine decay model based on two competitive reactions

Another hypothesis (Clark and Sivaganesan 2002) is based on two competitive reactions (fast and slow) that describe chlorine decay in raw and finished water can be given as



Where  $C_{A1}$  is free chlorine residual reacting with the collection of rapidly reacting components;  $C_{B1}, C_{A2}$  are free chlorine residual reacting with the collection of more slowly reacting components  $C_{B2}, P_1$  and  $P_2$  are collection of by-products of the two reactions; and  $a_1, b_1, p_1, a_2, b_2$  and  $p_2$  are stoichiometric coefficients. These equation can, be used to quantify the fraction of initial chlorine being utilized by fast and slow reacting components. The governing equations for second order kinetics for above reactions are

The expressions given below, describe the change in  $C_{A1}$  and  $C_{A2}$  with time (Clark and Sivaganesan 1998)

$$C_{A1}(t) = \frac{Cl_{01}(1-R_1)}{1-R_1 e^{(1-R_1)K_1 t}} \quad (18)$$

$$C_{A2}(t) = \frac{Cl_{02}(1-R_2)}{1-R_2 e^{(1-R_2)K_2 t}} \quad (19)$$

Where  $Cl_{01}$  and  $Cl_{02}$  are initial concentrations of  $C_{A1}$  and  $C_{A2}$ ,  $C_{A1}(t)$  and  $C_{A2}(t)$  are change in the concentration of  $C_{A1}$  and  $C_{A2}$  with time;  $K_1, K_2, R_1, \text{ and } R_2$  are parameter in equation (18) and (19); and  $t$  represents time. The total initial chlorine concentration at time zero is

$$Cl_0 = Cl_{01} + Cl_{02} \quad (20)$$

Where  $Cl_0$  is total initial chlorine residual.  $Cl_{01}$  and  $Cl_{02}$  are the initial chlorine concentration of chlorine reacting in equation (16) and (17) then

$$Cl_{02} = Cl_0 - Cl_{01} \quad (21)$$

The equation for the complete reaction is the sum of equation (18) and (19) or

$$Cl(t) = \frac{Cl_{01}(1-R_1)}{1-R_1e^{(1-R_1)K_1t}} + \frac{(Cl_0 - Cl_{01})(1-R_2)}{1-R_2e^{(1-R_2)K_2t}} \quad (22)$$

Given that  $Cl_0$  is know and that  $Cl_{01}, K_1, K_2, R_1, \text{ and } R_2$  are unknown, equation (22) yield a five parameter equation:

$$Cl(t) = \frac{Cl_0Z(1-R_1)}{1-R_1e^{(1-R_1)K_1t}} + \frac{Cl_0(1-Z)(1-R_2)}{1-R_2e^{(1-R_2)K_2t}} \quad (23)$$

Where  $Cl(t)$  = residual chlorine at  $t$  hours;  $Z = (Cl_{01}/Cl_0)$ ; and  $K_1, K_2, R_1, \text{ and } R_2$  are unknown parameters. All the model parameters are positive and  $Z$  cannot be larger than 1.

## Material and method

A different approach of multiple reaction constituent model for the chlorine reactions with fast reacting agents and slow reacting agents can be represented by the following equations



Where  $P_1$  and  $P_2$  are byproducts of the reactions between chlorine with fast reacting agents and slow reacting agents respectively

According to second order kinetics for both above reactions the concentration decay equations can be given by s

$$\frac{dc_1}{dt} = K_1cc_1 \quad (26)$$

$$\frac{dc_2}{dt} = K_2cc_2 \quad (27)$$

$$\frac{dc}{dt} = \frac{dc_1}{dt} + \frac{dc_2}{dt} = -c(K_1c_1 + K_2c_2) \quad (28)$$

where  $c_1, c_2$  and  $c$  are fast reacting agents, slow reacting agents and chlorine concentration respectively.

According to mass balance at time  $t$ , the amount of chlorine concentration is subtracted from fast reacting agents and slow reacting agents is equal to total chlorine concentration. Therefore, if  $c_0$  represents initial chlorine concentration  $c_{10}$  and  $c_{20}$  are initial concentration of fast reacting agents and slow reacting agents respectively and if  $a$  and  $b$  are the concentrations of fast reacting agents and slow reacting agents that have reacted with chlorine at time  $t$ , then free chlorine in water is

$$c = c_0 - a - b \quad (29)$$

$$\frac{dc}{dt} = -\frac{da}{dt} - \frac{db}{dt} \quad (30)$$

$$c_1 = c_{10} - a \Rightarrow \frac{dc_1}{dt} = -\frac{da}{dt} \quad (31)$$

$$c_2 = c_{20} - b \Rightarrow \frac{dc_2}{dt} = -\frac{db}{dt} \quad (32)$$

Initial condition  $a = 0, b = 0$  at  $t = 0$

Using equations (29) to (32) in (26) and (27), we get

$$\frac{dx}{dt} = K_1(c_0 - a - b)(c_{10} - a) \quad (33)$$

$$\frac{dy}{dt} = K_2(c_0 - a - b)(c_{20} - a) \quad (34)$$

For solving equations (33) and (34)

$$\frac{dx}{dy} = \frac{K_1(c_{10} - a)}{K_2(c_{20} - b)} \quad (35)$$

Solutions of above differential equations with initial condition ( $t = 0, a = 0, b = 0$ ), are

$$a = c_{10} \left( 1 - \left( \frac{c_{20} - b}{c_{20}} \right)^{K_1/K_2} \right) \quad (36)$$

$$b = c_{20} \left( 1 - \left( \frac{c_{10} - a}{c_{10}} \right)^{K_2/K_1} \right) \quad (37)$$

Using equation (37) in (33) and integrating, yield

$$\int \frac{da}{\left( c_0 - a - c_{20} \left( 1 - \left( \frac{c_{10} - a}{c_{10}} \right)^\alpha \right) \right) (c_{10} - a)} = K_2 t + const \quad (38)$$

where  $\alpha = \frac{K_2}{K_1}$ .

Similarly using equation (36) in (34) and integrating, we get

$$\int \frac{db}{\left( c_0 - b - c_{10} \left( 1 - \left( \frac{c_{20} - b}{c_{20}} \right)^{1/\alpha} \right) \right) (c_{20} - b)} = K_1 t + const \quad (39)$$

Since  $K_1 \gg K_2$  (i.e. chlorine decay rate constant for fast reacting agent is much larger than chlorine decay rate constant for slow reacting agent) so initially for a short time period,  $K_2 \rightarrow 0$  therefore  $\alpha = \frac{K_2}{K_1} \rightarrow 0$ . Then

solutions of equations (38) and (39) can be given by

$$a = \frac{c_0 c_{10} (e^{-\beta} - 1)}{c_{10} e^{-\beta} - c_0} \quad (40)$$

where  $\beta = (c_0 - c_{10}) K_1 t$

$$b = \frac{c_{20}(c_0 - c_{10})(1 - e^{-\gamma})}{c_0 - c_{10} - c_{20}e^{-\gamma}} \quad (41)$$

where  $\gamma = (c_0 - c_{10} - c_{20})K_2t$ .

The solutions given by equation (40) and (41) satisfy the final condition that as  $t \rightarrow \infty$ ,  $a = c_{10}$ ,  $b = c_{20}$ . Finally free chlorine concentration in water can be given by

$$c = c_0 - a - b$$

For the time  $t > t_1$ , assuming that the reactions in the water due to fast reacting agent is over and only the slow reacting agent is remaining. In this case the free chlorine in the water can be calculated as given by Clark [2].

### One Dimensional first order Chlorine Decay Model

The unsteady-state mass conservation equation for the concentration of the chlorine in reservoir considering diffusion of chlorine in x direction and first order chlorine decay, can be written as

$$\frac{dc}{dt} = D \frac{d^2c}{dx^2} - Kc \quad (42)$$

where  $D$  is diffusion coefficient,  $K$  is the chlorine decay rate constant for bulk flow ( $s^{-1}$ ).

The initial and boundary conditions are

$$c = 0, t = 0, x \geq 0 \quad (43.i)$$

$$c = c_0, t > 0, x = 0 \quad (43.ii)$$

$$c = 0, x \rightarrow \infty, t \geq 0 \quad (43.iii)$$

The solution of equation (42) together with initial boundary conditions (43.i) to (43.iii), is

$$c = \frac{c_0}{2} \left[ \exp\left(-x\sqrt{K/D}\right) \operatorname{erfc}\left(\frac{x}{2\sqrt{Dt}} - \sqrt{Kt}\right) + \exp\left(x\sqrt{K/D}\right) \operatorname{erfc}\left(\frac{x}{2\sqrt{Dt}} + \sqrt{Kt}\right) \right] \quad (44)$$

### Two Dimensional diffusion and first order Chlorine Decay Model

The mass conservation equation considering diffusion in x and y directions and first order chlorine decay in reservoir can be written as

$$\frac{\partial c}{\partial t} = D_x \frac{\partial^2 c}{\partial x^2} + D_y \frac{\partial^2 c}{\partial y^2} - Kc \quad (45)$$

where  $D_x$  is diffusion coefficient in x direction,  $D_y$  is diffusion coefficient in y direction,  $K$  is the chlorine decay rate constant for bulk flow ( $s^{-1}$ )

The initial boundary conditions are

$$c = 0, t = 0, x \geq 0, y \geq 0 \quad (46.i)$$

$$c = c_0, t > 0, x = 0, y = 0 \quad (46.ii)$$

$$c = 0, x \rightarrow \infty, y \rightarrow \infty, t \geq 0 \quad (46.iii)$$

where  $c_0$  is initial concentration

Introducing a new space variable

$$X = x + y \sqrt{\frac{D_y}{D_x}} \quad \text{in equation (43), (44.i) and (44.ii), we get}$$

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial X^2} - Kc \quad (47)$$

$$\text{Where } D = D_x \left( 1 + \frac{D_y^2}{D_x^2} \right)$$

The initial and boundary condition become

$$c = 0, t = 0, X \geq 0 \quad (48.i)$$

$$c = c_0, t > 0, X = 0 \quad (48.ii)$$

$$c = 0, X \rightarrow \infty, t \geq 0 \quad (48.iii)$$

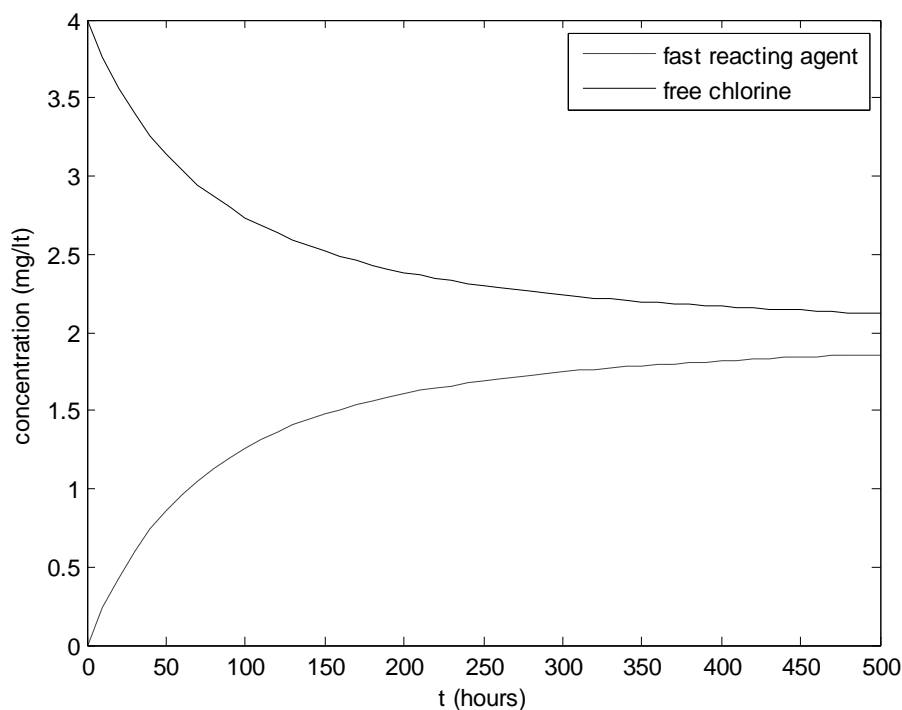
Now this problem is similar to previous problem (i.e. equation (42) and (43) ) and therefore its solution can be given as

$$c = \frac{c_0}{2} \left[ \exp\left(-X\sqrt{K/D}\right) \operatorname{erfc}\left(\frac{X}{2\sqrt{Dt}} - \sqrt{Kt}\right) + \exp\left(X\sqrt{K/D}\right) \operatorname{erfc}\left(\frac{X}{2\sqrt{Dt}} + \sqrt{Kt}\right) \right] \quad (49)$$

$$\text{where } X = x + y\sqrt{\frac{D_y}{D_x}}$$

## RESULTS AND DISCUSSION

As the free chlorine for the multiple reactions constituent model of chlorine with fast reacting agents and slow reacting agents is given by  $c = c_0 - a - b$  where a is the concentration of fast reacting agents and b is the concentration of slow reacting agents and which are given by equation (40) and (41).



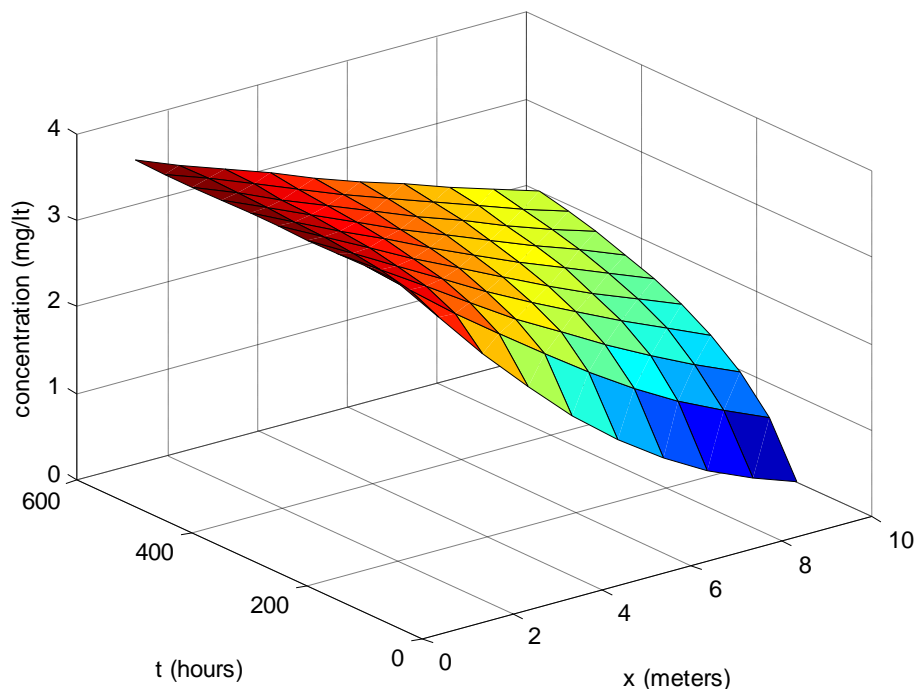
**Fig.1 Variation of concentrations of free chlorine and fast reacting agent with time t ( $c_0 = 4$ ,  $c_{10} = 2$ ,  $c_{20} = 1$ )**

The numerical results for free chlorine c and for concentration of fast reacting agents a are shown in fig.1 with initial chlorine concentration  $c_0 = 4$ , final concentration  $c_{10} = 2$  for fast reacting agents and  $c_{20} = 1$  for slow reacting agents. It is clear from the figure that from  $t = 0$  to  $t = 100$  concentration of free chlorine decreases rapidly while

from  $t = 150$  to  $t = 350$ ,  $c$  decreases slowly and after that change in  $c$  with time is very small. This is due to fact that the concentration of fast reacting agents increases very fast in starting period after that it becomes slow and finally it becomes almost constant.

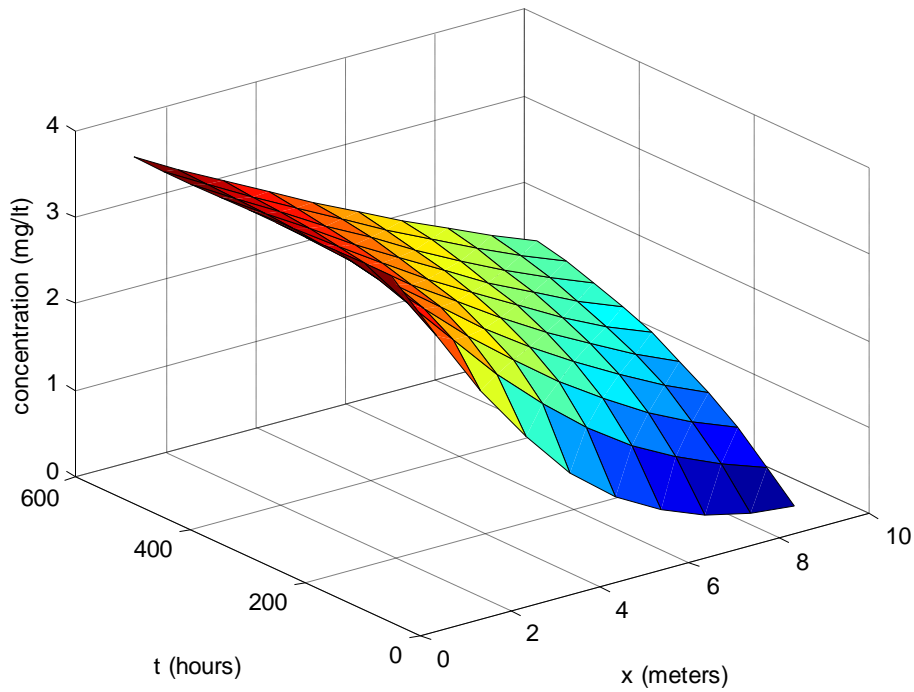
The free chlorine concentration for first order decay model together with diffusion in  $x$  direction is given by equation (44) and the free chlorine concentration for first order decay model with diffusion in  $x$  and  $y$  both directions is given by equation (49). Both the results given by equation (44) and (49) appear similar except

$X = x + y \sqrt{\frac{D_y}{D_x}}$  in equation (49). Thus we have plotted the figure (2) to (4) only for equation (44).



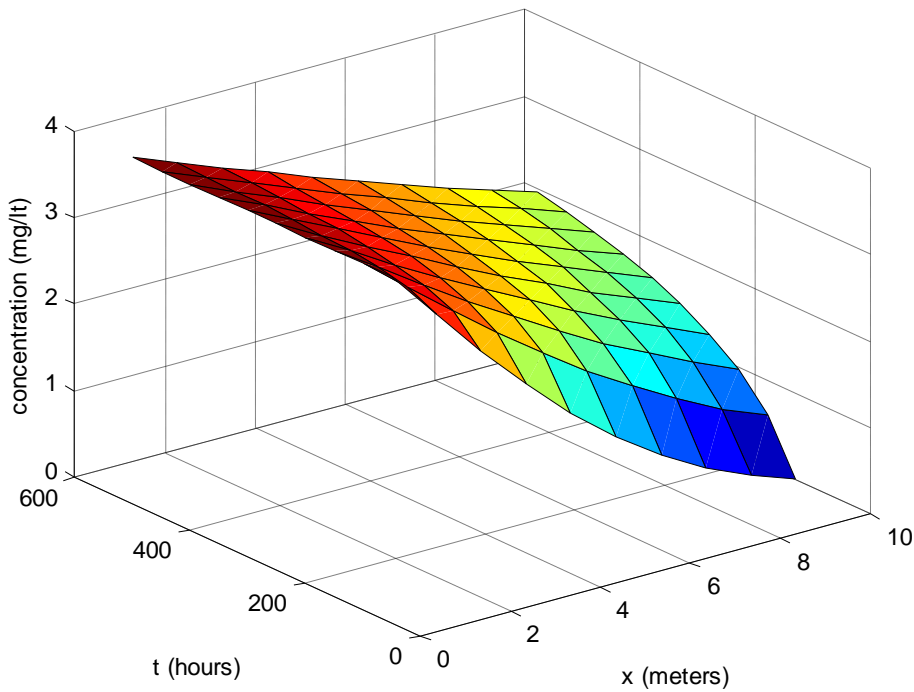
**Fig.2 Variation of chlorine concentration with distance and time ( $c_0 = 4$ ,  $K = 0.0001$ ,  $D = 0.003$ )**

In fig.2 we have taken initial chlorine concentration  $c_0 = 4$  at  $x = 0$  and  $t = 0$ . It is also assumed that at  $x = 0$  chlorine is continuously injected so as to maintain boundary condition (43.ii). We have observed that chlorine concentration decreases as the distance in  $x$  direction increases which is a true fact. The numerical value of chlorine concentration  $c = 3.16$  (at  $t = 50$  and  $x = 1$ ) while  $c = 0.07054$  (at  $t = 50$  and  $x = 10$ ). From these results we see that at  $x = 10$  meter the chlorine concentration is below the limit for safe drinking water in reservoir. Therefore to maintain the safe limit of chlorine in drinking water within stipulated time chlorine must be injected at more than one point in a reservoir. The effect of diffusivity on the free chlorine in water can be observed by comparing fig.2 and fig.3. Both fig.2 and fig.3 appear to be similar but when we see the numerical value the effect of diffusivity on free chlorine is quite clear.



**Fig.3 Variation of chlorine concentration with distance and time ( $c_0 = 4$ ,  $K = 0.0001$ ,  $D = 0.00144$ )**

In fig.2 for diffusivity  $D = 0.003$ ,  $c = 0.40400016$  (at  $t = 50$  and  $x = 10$ ) while for  $D = 0.000144$  (in fig.3),  $c = 0.7057$  (at  $t = 50$  and  $x = 10$ ). Clearly chlorine concentration decreases when diffusivity increases, which is due to fact that more mixing takes place when diffusivity increases.



**Fig.4 Variation of chlorine concentration with distance and time ( $c_0 = 4$ ,  $K = 0.0002$ ,  $D = 0.003$ )**

To see the effect of chlorine decay rate constant  $K$ , we have plotted fig.4 for  $K = 0.0002$  and fig.1 for  $K = 0.0001$  keeping other parameters same. We have observed that for  $K = 0.0001$ ,  $c = 0.400016$  (at  $x = 10$  and  $t = 50$ ) while for  $K = 0.0002$ ,  $c = 0.398646$  (at  $x = 10$  and  $t = 50$ ). This shows that free chlorine in water reservoir decreases as the chlorine decay rate constant increases and which is a true fact.

## Conclusion

In this paper we have shown the effect of fast reacting agents and slow reacting agents with second order decay rate constant on the free chlorine in water. We have considered the models with diffusion term in  $x$  as well as in  $y$  direction together with first order decay rate constant and found that free chlorine changes not only with time but with distance also. It is found that injection of chlorine at one point in a reservoir is not sufficient to maintain the safe limit of the chlorine in drinking water. The model can be used to locate the point for booster chlorination.

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