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Modeling of Ozone as a Disinfectant of Indicators Bacteria in the Drinking Water

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ABSTRACT: Background: Ozone is considered one of the most effective disinfectants that can inactivate resistant pathogenic microorganisms in which conventional disinfectant such as chlorine and chlorine dioxide fail due to its strong biocidal oxidizing property. **Objective:** The study aimed at modeling of ozone as a disinfectant of indicators bacteria in the drinking water. **Methods:** This work involved a series of batch experiments with raw water, taken from the intake of El-Nozha Water Purification Plant, Alexandria governorate. The ozone doses applied in this study were 1.2, 2.2, 3.4, and 4.3 mg/l. The disinfected effluent was collected at 5, 10, 20, and 30 min. The indicator microorganisms HPC bacteria, total coliforms, faecal coliforms and *Streptococcus faecalis* were examined before and after the ozonation. **Results:** The optimum ozone dosage applied in raw water to achieve 90% reduction of the indicator microorganisms was 2.5 mg/l. The optimum contact time to achieve 90% reduction of the indicator microorganisms in raw water was 8 min. By applying of ozone as post disinfection on filtered water, the reduction percent of the indicator microorganisms were 100%. This study estimated that the ozone cost was 1.76 piasters/m^3**. Conclusion and Recommendations:** Ozone as a primary disinfection must be applied on raw water to reduce the formation of THMs due to pre-chlorination of raw water at present, in addition to effective killing power of ozone onto microorganisms that will improve water quality.

Keywords: Disinfection; Faecal Coliforms; HPC Bacteria; Ozone; Streptococcus Fecalis; Total Coliforms; Water Purification Plant.

INTRODUCTION

Drinking water contaminated with pathogenic microorganisms may be a major source of infectious diseases. The conventional water treatment processes including pre-chlorination, coagulation, flocculation, sedimentation and filtration

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that remove many microorganisms from the water along with the suspended solids. But these processes are not sufficient to ensure the complete removal of pathogenic bacteria or viruses. To accomplish this, the final treatment process in water treatment plant is disinfection.[(1)]

The primary purpose of disinfecting raw water is to destroy and eliminate pathogenic organisms responsible for water borne-diseases. Monitoring of microorganisms in full-scale systems enables to assess contaminations in the raw water and the improvement through water treatment.[(2)]

There are several chemical oxidants used as disinfectants. Chlorine is the most commonly used disinfectant, but it reacts with organic matter present in most water sources to form chlorinated compounds, primarily trihalomethans (THMs). Most trihalomethans are of public health concern that (THMs) may cause cancer to humans.[(3)] Consequently, substantial efforts have been made to investigate the use of alternative disinfectants, in particular ozone.

Ozone is considered one of the most effective disinfectants that can inactivate resistant pathogenic microorganisms such as protozoa e.g. (Giardia and *Cryptosporidium parvum* oocyst) in which conventional disinfectant such as chlorine and chlorine dioxide fail due to its strong biocidal oxidizing property. Ozone is very effective against bacteria. Studies have shown the effect of small concentrations of dissolved ozone on *E. coli* and *Legionella pneumophila*.[(4)]

It has been observed that for a given concentration of a disinfectant, the longer the contact time, the greater the kill. The kinetics of disinfection follows the time rate of kill described by Chick's law of disinfection $\frac{dN}{dt} = - kN_t$

This law states that number of organisms destroyed per unit time is proportional to, the number of organisms remaining, where; N_t is number of organisms at time t, k is rate constant. Departures from Chick's law are common. Rates of kill may increase or decrease with time rather than remain constant with time.[5] To formulate a valid relationship for the kill of organisms under a variety of conditions an assumption made is that:

$$\ln \frac{N_t}{N_o} = -kt^m \quad \text{Where (m) is a constant.}$$

So, the purpose of this study was to set up modeling of ozone as a disinfectant of indicators bacteria in the drinking water.

MATERIAL AND METHODS

The present study was performed at the High Institute of Public Health laboratory, Alexandria University.

Study setting:

Ozonation experiments were performed with raw water taken from EL-Nozha Water Purification Plant in Alexandria governorate.

Sampling and analyses:

Grab samples of raw water (un-disinfected water) and disinfected water were collected for microbiological analysis. Samples were analyzed according to the Standard Methods for the Examination of Water and Wastewater. [6] The indicator microorganisms HPC bacteria, total coliforms, faecal coliforms and *Streptococcus faecalis* were examined before and after the ozonation.

All the experiments were conducted at temperature 23-25°C. The disinfected effluent was collected at 5, 10, 20, and 30 min in a sterile glass stopper bottle. The ozone dosage was varied by changing the adjustment of the ozone output regulator. The applied ozone doses used in this study were 1.2, 2.2, 3.4, and 4.3 mg/l.

Experimental setup:

A complete ozonation system was setup as shown in figure (1). The experimental apparatus consisted of the following components: air pump, air dryer, ozone generator, cylindrical ozone-water contactor (conducted by a diffusing bubble air stone).Ozone was generated in a gas phase by passing dry clean air through an ozone generator. The ozone gas was applied at the bottom of the reactor, where it bubbled through a diffusing air stone and moved upwards through the reactor.

Ozone-water contactor was designed as a semi-batch reactor, batch reactor with respect to the volume of water treated, and continuous-flow reactor with respect to ozone gas fed. The reactor was constructed from Pyrex-glass cylinder of 7.5 cm internal diameter and 40 cm in length. The top of the reactor was closed with a stopper. Three openings in the stopper of 8 mm were fitted with hollow glass tubing. The tubing used to transport the ozone was made of ozone resistant tubing of 6 mm. The first hollow glass tube allow ozone to be introduced and bubbled into the water sample by passing through a diffusing stone at the bottom of the reactor. The second opening tube allow the water sample withdrawal in which the end of the tube reached the middle of the reactor, while the third one allow the excess gases to be vented out.

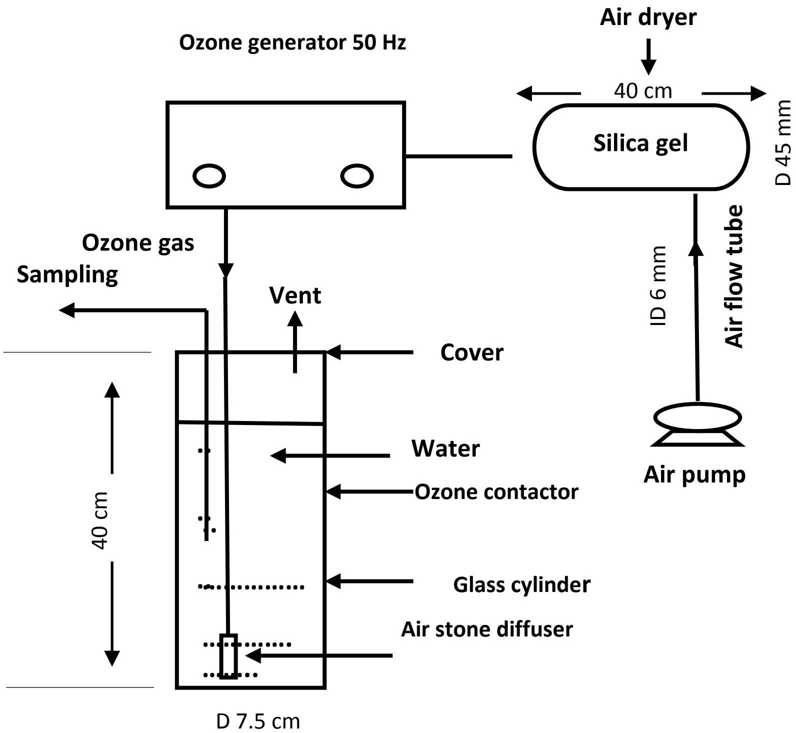

Figure (1): The Experimental Setup for Ozonation Experiments

A pump was used to deliver air. The flow rate of the air was measured and held constant all over the experiments. A graduated cylinder (1liter) was completely filled with water and inverted up-side down in a basin filled with water. The air delivered by the pump was introduced to the inverted graduated cylinder filled with water from down so air bubbles moved upwards. In the present study the flow rate conducted in all the experiments was approximately 0.5 l / min.

RESULTS AND DISCUSSION:

<u>A) Effect of ozone dosages as pre-disinfectant on indicator bacteria:</u>

Table (1) illustrates the effect of ozone dosages on the reduction of HPC bacteria, total coliforms, faecal coliforms and streptococcus faecalis in raw water from El Nozha Water Purification Plant. It is clear that the contact time is inversely correlated with ozone dose, and the incomplete removal of indicator bacteria was due to large number of bacteria (T.C) in raw turbid water.

Table (1): Effect of ozone dosages on the reduction of the indicator microorganisms in raw water from the intake of El Nozha Water Purification Plant, Alexandria governorate (2008)

Contact time (min)	Ozone dosages (mg/l)	Effluent HPC Bacteria		Effluent total coliforms		Effluent faecal coliforms		Effluent streptococcus faecalis	
		Geometric mean (CFU/ml)	Reduction%	Geometric mean /100 ml	Reduction%	Geometric mean /100 ml	Reduction%	Geometric mean /100 ml	Reduction%
0*	0	9786	0	2787	0	951	0	330	0
5	1.2	4230	56.78	1138	59.18	314	66.9	340	55.23
5	2.2	2630	73. 12	751	73.07	219	76.9	215	71.67
5	3.4	972	90.07	219	92.14	103	90	73	90.37
5	4.3	527	94.62	165	94.09	58	93.9	55	92.78
10	1.2	2380	75.68	686	75.37	208	78.2	215	71.72
10	2.2	1448	85.20	411	85.24	154	83.8	133	82.49
10	3.4	606	93.80	160	94.27	59	93.7	50	93.46
10	4.3	377	96.15	129	95.36	36	96.2	18	97.69
20	1.2	1472	84.96	402	85.56	136	85.7	151	80.11
20	2.2	678	93.07	223	91.99	86	90.9	98	87.10
20	3.4	436	95.55	121	95.67	36	96.1	40	94.79
20	4.3	327	96.66	96	96.55	8	99.1	11	98.54
30	1.2	1211	87.62	291	89.58	111	88.3	131	82.81
30	2.2	576	94.11	197	92.95	72	92.4	74	90.27
30	3.4	386	96.06	105	96.25	18	98.1	34	95.49
30	4.3	298	96.95	86	96.90	6	99.4	9	98.85

*Raw water samples without disinfections.

Physical analysis of influent raw water:

Turbidity for influent raw water ranged from (7 to 13) NTU with average of 10 NTU.

Temperature for influent raw water ranged from (22 to 25) Cº with average of 23 Cº.

pH for influent raw water ranged from 7.7 to 8.5.

$$\% \text{ Reduction} = \frac{\text{Influent} - \text{Effluent}}{\text{Influent}} \times 100$$

i) Mathematical model for reduction of indicator bacteria using ozone:

1. HPC Bacteria:

Figure (2) shows the relationship between the reduction percentage of HPC bactria and ozone dosages. As shown on the figure, the % reduction of HPC bacteria was a function of ozone dosages according to the mathematical polynomial model.

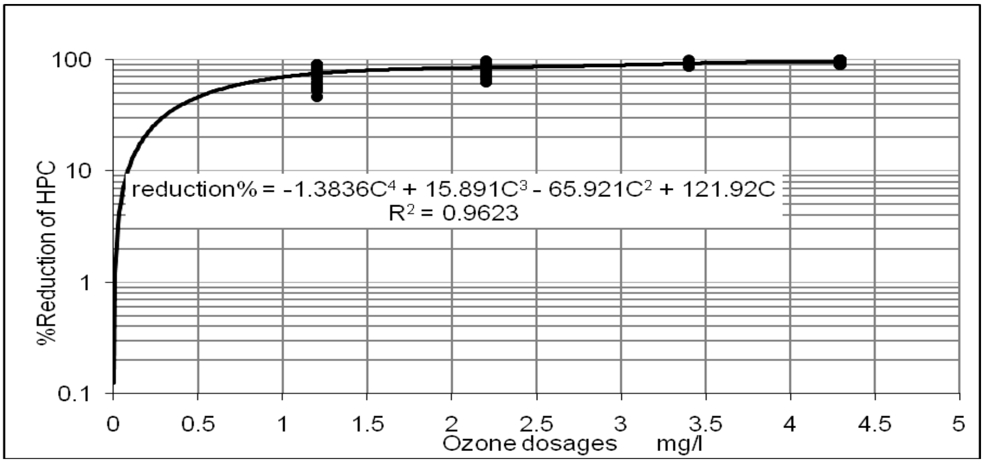

Figure (2): Optimum Ozone Dosage for Death of HPC Bacteria

2. Total Coliforms:

Figure (3) shows the relationship between the reduction percentage of total coliforms and ozone dosages.

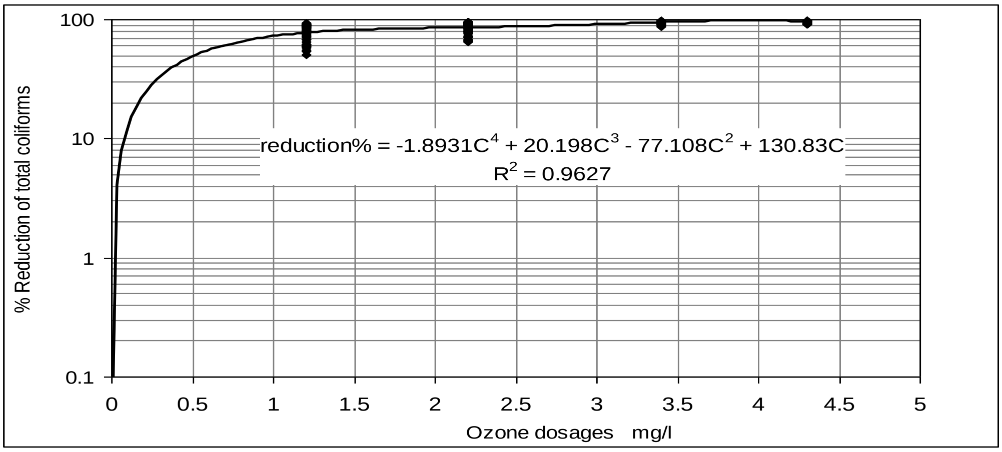

Figure (3): Optimum Ozone Dosage for Death of Total Coliforms

3. Faecal Coliforms:

Figure (4) shows the relationship between the reduction percentage of total coliforms and ozone dosages.

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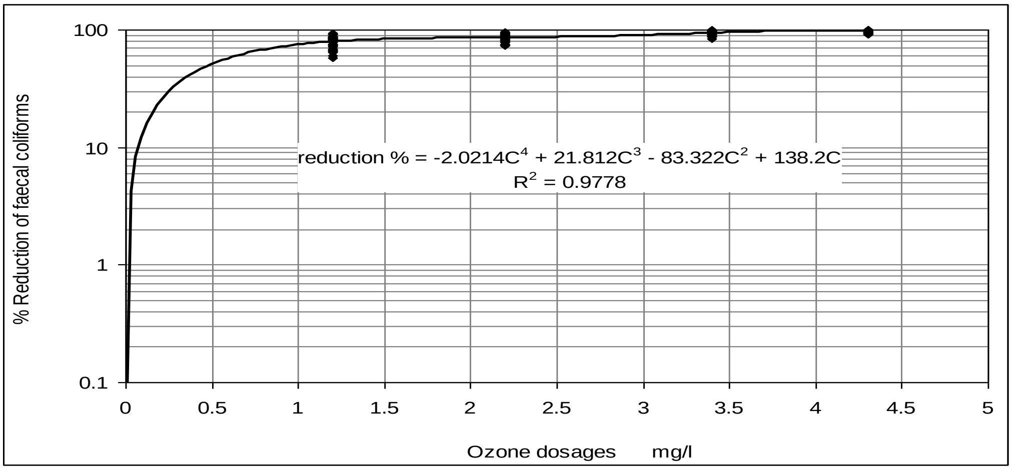

Figure (4): Optimum Ozone Dose for Death of Faecal Coliforms

4. Streptococcus faecalis

Figure (5) shows the relationship between the reduction percentage of *Streptococcus Fecalis* and ozone dosages.

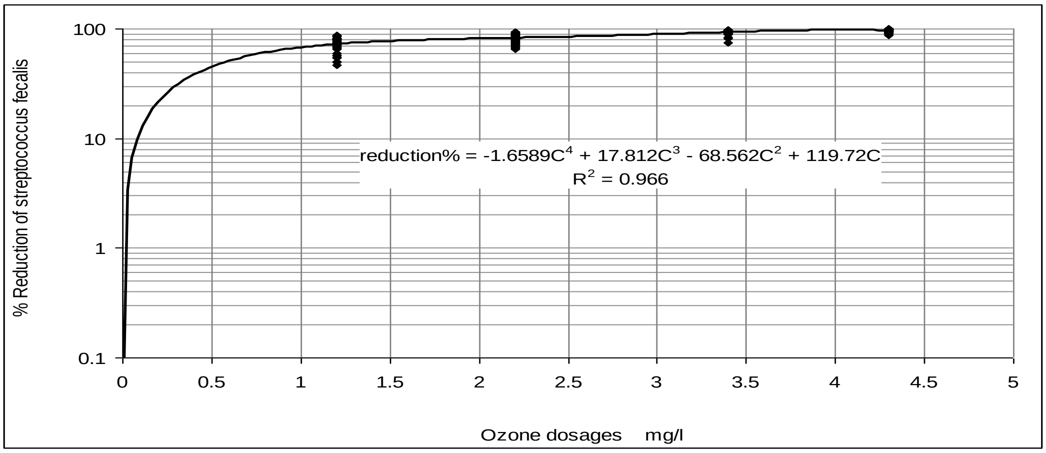

Figure (5): Optimum Ozone Dose for Death of *Streptococcus Fecalis*

The figures show that the reduction percentage of indicators bacteria increased rapidly by increasing ozone dosages, followed by slight increase in the percentage reduction by increasing ozone dosages.

From these results, it was observed that the optimum ozone dosage to achieve 90% reduction of HPC bacteria and total coliforms was 2.2 mg/l min. while, for faecal coliforms and Streptococcus Fecalis was 1.8, and 2.5 mg/l respectively.

Disinfection mechanism of bacteria by ozone is attributed to an oxidation reaction, through the addition of an atom of oxygen, and by ozonolysis, which enables it to act on double bond by fixing the complete ozone molecule on the double bond atoms (acting on proteins, or enzymes). The first site to be attacked appears to be the bacterial membrane either through the glycoproteins or glycolipids or through certain amino acids such as typtophan. In addition, ozone disrupts enzymatic activity of bacteria by acting on the sulfhydryl groups of certain enzymes. Beyond the cell membrane and cell wall, ozone may act on the nuclear material within the cell. Ozone has been found to affect both purines and pyrimidines in nucleic acids.(7)

It is observed that when the concentration of ozone is increased, the time required to obtain a certain level of disinfection (% reduction) is less at higher ozone concentrations. These results were in accordance with other workers.(8)

Amirsaradi *et al* studies were conducted to evaluate the effect of pre-ozonation on disinfection, disinfection by-product precursors and water quality in a direct filtration water treatment system. Disinfection parameters including total coliforms, faecal coliforms and heterotrophic plate count were investigated. Total organic carbon (TOC), trihalomethanes (THMs), total organic halides (TOX), filtered water turbidity and colour were also evaluated. It was found that advanced pre-oxidation processes (ozonation) significantly increase the level of disinfection of raw water. Removal of total trihalomethanes and total organic halides precursors improved with ozonation, compared to no oxidation treatment in direct filtration and/or in conventional water treatment. All coliforms (total and faecal) were completely destroyed by ozonation alone. (9)

ii) Mathematical model to evaluate the effect of contact time on disinfection efficiency:

Experiments were also conducted to determine the effect of increasing contact time (5, 10, 20 and 30 min) on the reduction of HPC bacteria, total coliforms, faecal coliforms and streptococcus fecalis in raw water taken from the intake of EL Nozha Water Purification Plant. Four ozone dosages (1.2, 2.2, 3.4 and 4.3 mg/liter) were tested.

1. HPC bacteria:

Table (1) and figure (6) show the

reduction of HPC bacteria with contact time. Almost, no additional reduction was observed when the contact times were increased to 10 and 30 min.

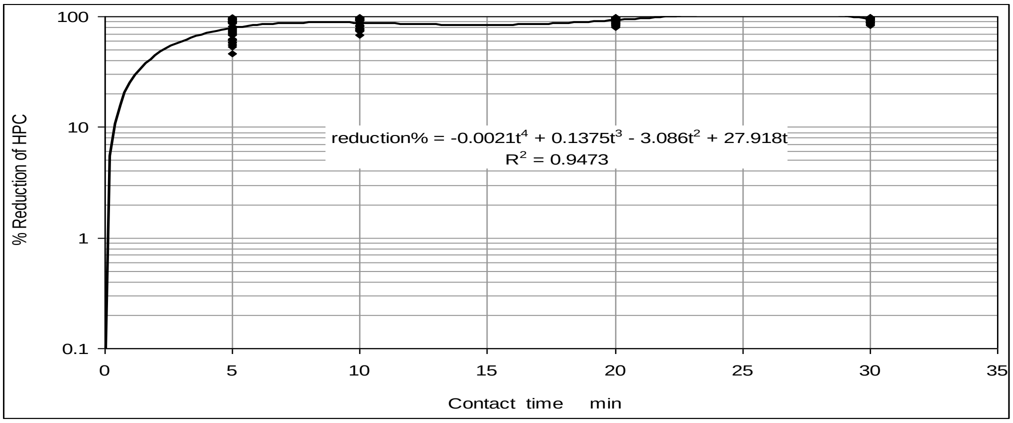

Figure (6): Optimum Contact Time for Death of HPC Bacteria

2. Total Coliforms:

Figure (7) shows the relationship between the reduction percentage of total coliforms and contact time. It is obvious from the figures, the reduction percentage reached its maximum (over 90%) rapidly at time 8 min, followed by a slight decrease in the reduction percentage, and again a slowly increase in the reduction percentage by increasing contact time.

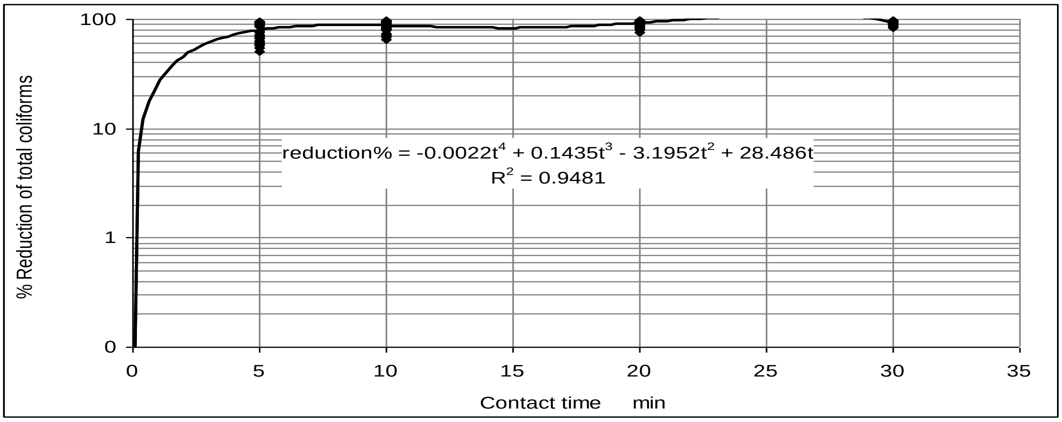

Figure (7): Optimum Contact Time for Death of Total Coliforms

3. Faecal Coliforms:

Figure (8) shows the relationship between the reduction percentage of Faecal Coliforms and contact time.

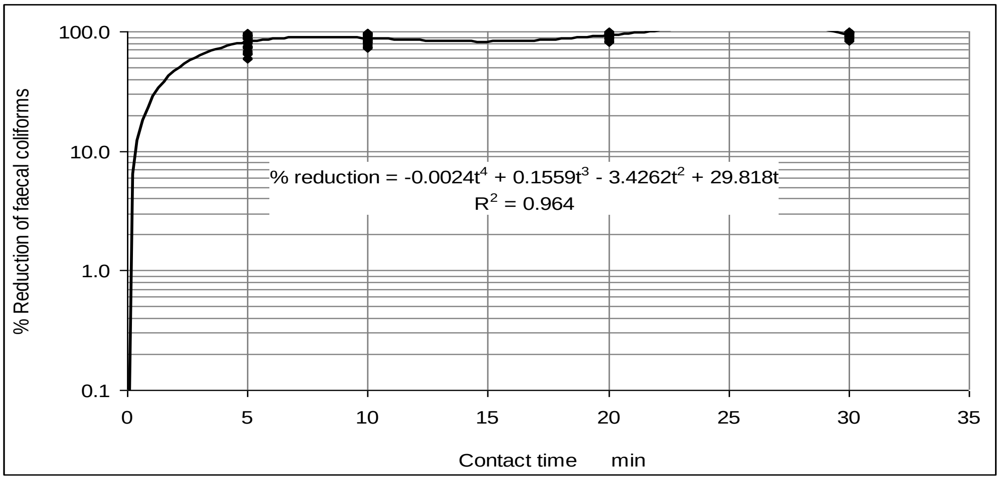

Figure (8): Optimum Contact Time for Death of Faecal Coliforms

4. Streptococcus faecalis

Figure (9) shows the relationship between the reduction percentage of Streptococcus Faecalis and contact time.

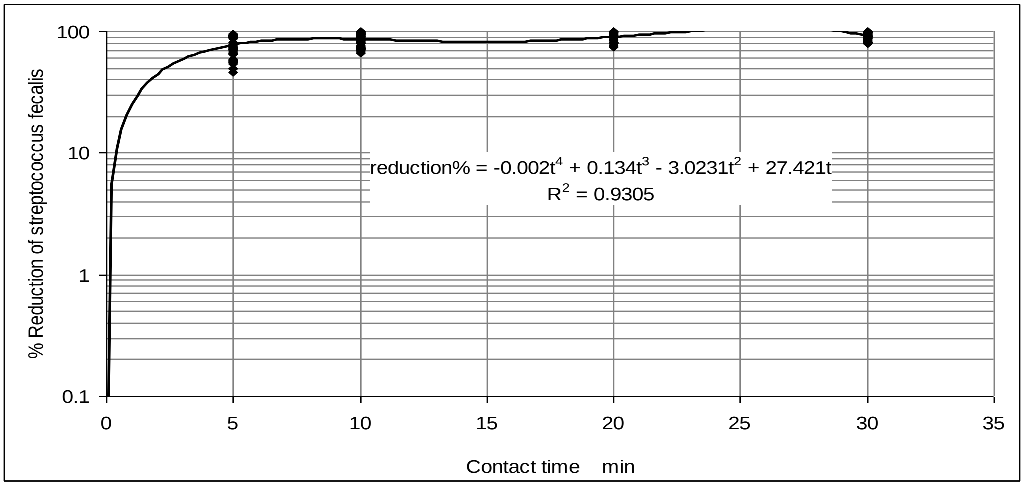

Figure (9): Optimum Contact Time for Death of *Streptococcus Faecalis*

From these results, it was observed that the optimum contact time to achieve 90% reduction of HPC bacteria, total coliforms and streptococcus fecalis was 8 min. while the optimum contact time to achieve 90% reduction of faecal coliforms was 6 min.

Increasing the contact time from 5 to 30 min improved HPC bacteria, total coliforms, faecal coliforms and *Streptococcus Fecalis* reduction.

Xu *et al* results showed that the time has no impact on the performance of faecal coliforms or E.coli disinfection, 2 min provides the same disinfection as 10 min. These results had major consequence for the design of ozone contactor for raw water disinfection demonstrating that no long contact time chamber is necessary.(10)

Blank *et al* results was that the reduction in faecal coliforms achieved in the ozone contactor were reported 99% - 99.99% by applying ozone dose of 6 mg/L and contact time 10 min.(11)

In the present study, the failure to achieve high levels of disinfection e.g. 99.9% can be attributed to some protection afforded by turbidity present in raw water; the average value of turbidity of raw water was 10 nephelometric turbidity units. This explanation is consistent with the fact that protection by solids may results in the diminution of microbial activation.

Foster *et al* found that turbidity levels of 5 nephelometric turbidity units provided protection of faecal coliforms when the ozone residual was ≤ 0.1. However no protection from disinfection was observed when the turbidity was ≤ 1 nephelometric turbidity units.(12)

Budde *et al* determined that turbidity was the most significant factor influencing faecal coliforms inactivation by ozone in waste water. The increased turbidity levels could have afforded the microorganism physical protection from disinfection.(13)

The results indicated that that the HPC bacteria was more resistant to ozone disinfection than faecal coliforms. These results agree with Wolfe *et al* results. The resistance of fecal streptococci to ozonation was greater than that of faecal coliforms. (14)

iii) Kinetics of disinfection

The following figures (10-13) illustrate the kinetics of disinfection of HPC bacteria, total coliform, faecal coliforms and streptococcus fecalis by applying various ozone dosages (1.2, 2.2, 3.4 and 4.3 mg/l) and contact time of (5, 10, 20 and 30 min). The following equation represents the kinetics of disinfection:

$$\mathrm{Ln}\,(N/N_0) = kt^m$$

Where: (N/N_0) is survival ratio of microorganisms, (N) is number of organisms at contact time (t) min, (N_0) is number of organisms before disinfection, (k) is kinetic rate constant min^{-1}, and (m) is kinetic parameter.

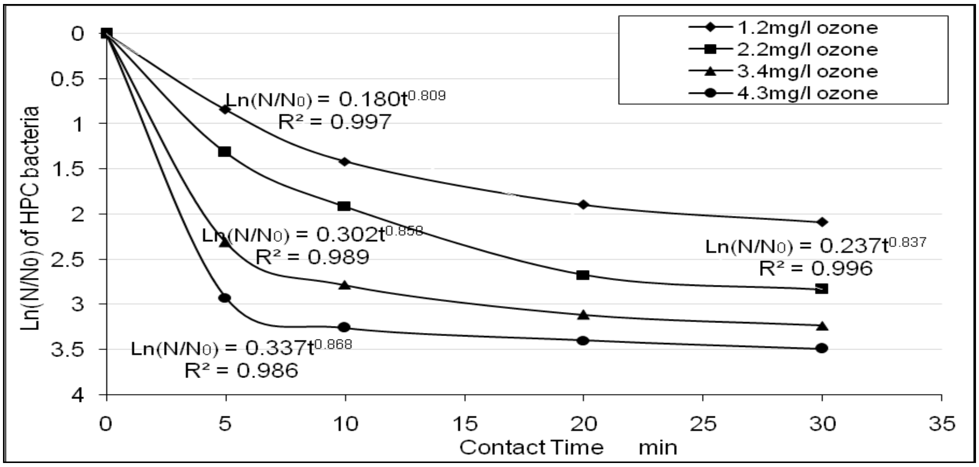

Figure (10): Kinetics of Death of HPC Bacteria by Applying Various Ozone Dosages

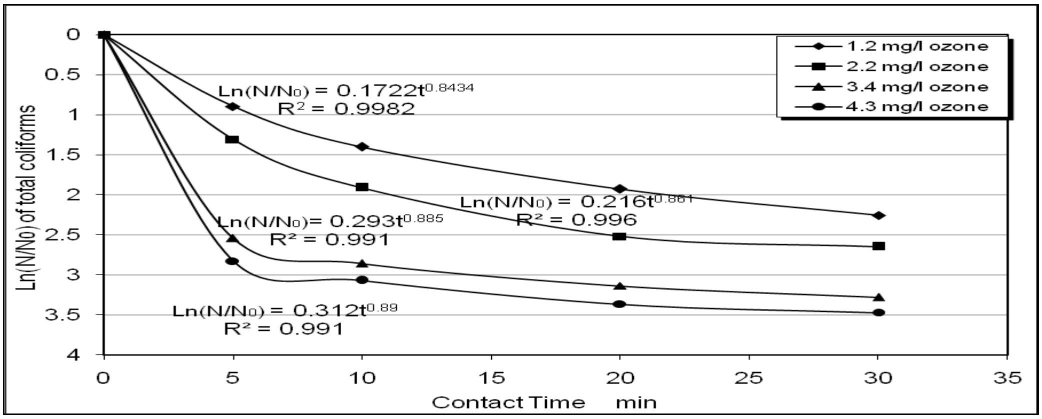

Figure (11): Kinetics of Death of Rate of Total Coliforms by Applying Various Ozone Dosages

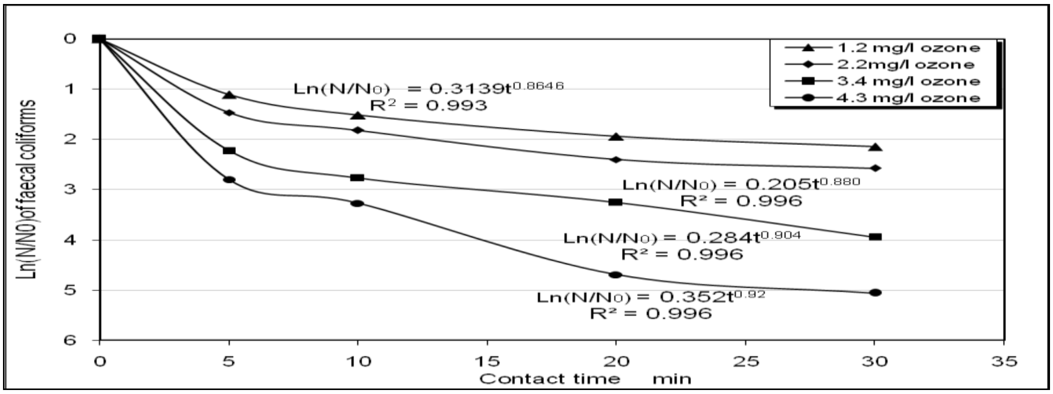

Figure (12): Kinetics of Death rate of Faecal Coliforms by Applying Various Ozone Dosages

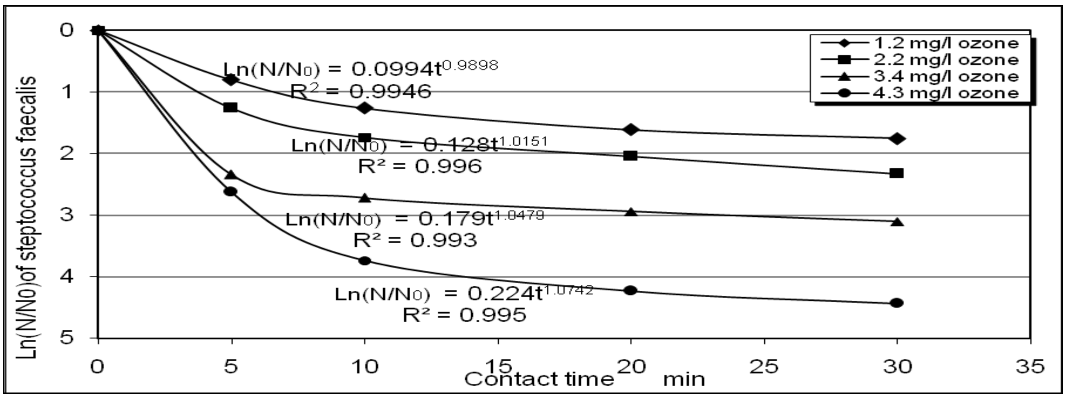

Figure (13): Kinetics of Death Rate of Streptococcus Faecalis by Applying Various Ozone Dosages

The results indicated that the disinfection kinetics were nonlinear, where rate of death declining with increasing contact time. In many cases, no additional death occurred beyond 10 min of exposure. At short contact time 10 min, the initial rate of death of bacteria was greatest, at longer contact time 30 min little additional of death is noticed while the bacteria continued to decline slightly.

It appeared that the ozone concentration was more important than contact time in determining the overall amount of death. These results are consistent with those obtained by a number of other researchers. [15]

B) Applying of ozone as post disinfection on filtered water

Table (2): Applying Ozone as Post Disinfectant on Filtered Water Taken from El-Nozha Water Purification Plant in Alexanderia Governorate, November 2008

Parameters	No. of samples	Filtered water		Reduction %
		Before ozone disinfection	After ozone disinfection	
HPC bacteria (CFU/ml)	5	80	1	98.5
Total coliforms (MPN/100ml)	5	74	N.D.	100
Faecal coliforms (MPN/100ml)	5	20	N.D.	100
Streptococcus fecalis MPN/100ml	5	20	N.D.	100

♦Pre-chlorination dose zero

By applying the optimum dosages of ozone (2.5 mg/l) and optimum contact times (8 min) on filtered water. It is clear that the reduction percent of the total coliforms, faecal coliforms, and streptococcus fecalis were 100% as shown in table (2). Choi *et al*, showed that it is more efficient to allocate the ozone disinfection process at the end stage of water treatment line, such as after filtration step in water treatment plant. [16]

Cost estimation of ozone

One pound ozone generated needs 10 KWh; (0.45 kg ozone generated needs 10 KWh)

1 KWh prices 32 piaster →10 KWh prices 3.2 L.E.

1 kg ozone prices 7.04 L.E.

Ozone dosage applied = 2.5 mg/L = 2.5×10^{-6} kg/ L

Ozone costs = price × ozone dosage applied = $7.04 \times 2.5 \times 10^{-6}$ L.E / L

= 17.6×10^{-6} L.E / L= 1.76 piaster/ m^3

The study estimated that the ozone cost was 1.76 piasters/m^3. Previous study carried out by Dyksen *et al* showed that ozone disinfection is the most cost-effective disinfection treatment. [17]

CONCLUSIONS

The ozone dosages had significant effect on the reduction of HPC bacteria, total coliforms, faecal coliforms, and *Streptococcus fecalis*. Greater reduction levels of all indicator organisms were achieved by increasing the applied ozone dosage. When the concentration of ozone is increased, the time required to obtain a certain level of disinfection (% reduction) decreased.

The optimum ozone dosage applied in raw water to achieve 90% reduction of HPC bacteria, total coliforms, faecal coliforms and *streptococcus fecalis* was 2.5 mg/l. The optimum contact time to achieve 90% reduction of HPC bacteria, total coliforms, faecal coliforms and streptococcus fecalis in raw water was 8 min.

Ozone as a primary disinfection must be applied on raw water to reduce the formation of THMs due to pre-chlorination of raw water at present, in addition to effective killing power of ozone onto microorganisms that will improve water quality.

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