Desalination and Water Treatment
www.deswater.com
2 (2009) 162–169
1944-3994/1944-3986

Prediction of trihalomethane formation in water distribution systems

H.A. Abu Qdais[a]*, N. Al-Abed[b], Z. Al-Ghazawi[a], F. Abdulla[a]

[a]*Civil Engineering Department,* [b]*Biosystems Engineering Department, Jordan University of Science and Technology, PO Box 3030, Irbid 22110, Jordan*
Fax: +962 2 709 5018; email: hqdais@just.edu.jo

Received 2 March 2007; Accepted 16 December 2008

ABSTRACT

Utilities involved in drinking water treatment are faced with a serious challenge in achieving the balance between pathogen destruction by chlorination and trihalomethanes (THMs) minimization below regulatory levels. Mathematical modeling offers an effective tool in cases where the prediction of THM formation is required. The main purpose of this study is to develop an empirical mathematical model that would predict the THM formation in water distribution systems on a laboratory scale. A parallel purpose of the study is to investigate, develop and test procedures for conducting water quality studies related to THM formation. Different types of regression models were investigated using backward elimination for the THM model parameters to select the optimum number of independent variables to be used. The Statistical Package for the Social Sciences backward regression revealed that there are five possible empirical models that predict the amount of total THMs formation as a function of various parameters such as, chlorine concentration, contact time, and temperature. The fifth model which only considers the free chlorine as independent variable, explains 80% ($R^2 = 0.8$) of the variability in total trihalomethanes (TTHM). This means that the best regression variable in predicting the TTHM is the free chlorine. The developed model serves as a basis for conducting further studies of THM formation on real scale water distribution networks.

Keywords: Chlorination; Trihalomethane; Modeling; Water distribution; Water quality; Chlorination by-product

1. Introduction

One of the major processes that is applied in the field of water treatment is the disinfection process. It is used to protect against microbial contamination of the water. Due to its high efficiency and the possibility of maintaining residual disinfectant in water distribution system with relatively low cost, chlorine is the most commonly used disinfectant in water treatment. However, chlorine interacts with organic matter that may be available in the raw water, such as humic and fulvic acids, to form disinfection by-products, such as trihalomethanes (THMs). THM compounds are the chlorinated and brominated derivatives of methane, namely, chloroform ($CHCl_3$), bromodichloromethane ($CHBrCl_2$), dibromochloromethane ($CHBr_2Cl$), and bromoform ($CHBr_3$).

The presence of THM compounds in chlorinated waters was first reported in the US by Bellar et al. in 1974 [1]. A later study by the US National Cancer Institute (1976) indicated that chloroform, the major component of THMs, is an animal carcinogen and, thus, is a suspected human carcinogen [2]. The carcinogenic effects of bromoform and bromodichloromethane were later reported [3,4]. The levels of THMs are currently limited by US EPA to 0.1 mg/L in distribution systems serving 10,000 people or more.

*Corresponding author.

Uyak [5] estimated the lifetime cancer risk and the hazard index of THMs through oral ingestion, dermal absorption, and inhalation exposure from tap water of 15 districts in Istanbul, Turkey. The most dominant THM compounds are chloroform, bromodichloromethane (BDCM), and dibromochloromethane (DBCM) in Istanbul tap water. The results indicated that the major pathway with higher cancer risk is the oral pathway. The researcher estimated that approximately 5 of the 8 million Istanbul residents could get cancer from the daily intake of tap water.

Water utilities are being forced to keep a sufficient amount of residual chlorine in the water distribution system to guard against microbial contamination. On the other hand, it is necessary to keep chlorine concentration to a minimum so as to decrease the THM formation potential. As such, the utilities involved in drinking water treatment are faced with a serious challenge in finding the balance between pathogen destruction and THM minimization as presented in Fig. 1.

Mathematical models for predicting water quality are considered an effective tool to evaluate water quality changes in water distribution system [6]. The effective use of these models would offer a number of benefits to the water industry. Such models would be useful in predicting water quality degradation problems, such as the THM formation.

The formation of THMs in drinking water depends on various water quality parameters and chlorination conditions. These include the total organic carbon level, the type of organic precursor, chlorination pH, temperature, bromide level, chlorine dosage, reaction time, and UV-254 absorbance [7]. However, analytical mechanistic-kinetic models of THM formation have not yet been developed mainly because of the complexity of the reactions of chlorine with organic materials in water. The consumption of chlorine in water occurs through complex consecutive and parallel reaction pathways. The rates that may describe the kinetics of these complex reactions are generally not well known [8]. As a result, description based on empirical reaction kinetics models is generally required.

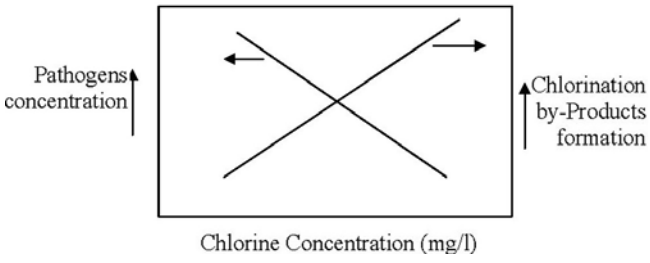

Fig. 1. Balance between disinfection and THM formation potential during chlorination process [4].

In order to describe the mechanisms of THMs formation and kinetics, several empirical models have been proposed. Lou and Chiang [9] studied the formation of trihalomethanes in a water distribution system in Taipei City (Taiwan) water distribution system. The study compared the measured values with the calculated values according to a model developed for water in a distribution system. They used a model of formation of TTHM which was developed earlier to predict the TTHM concentration. The model took into account dissolved organic carbon (DOC) concentration, chlorine dosage, temperature, pH, bromide concentration, contact time, and flow conditions of the piping system for drinking water. The applied model tended to overestimate the concentration of TTHM in the distribution system. As a result of the significant deviation between the predicted and measured values, the authors recommended not to apply their model in regulatory analysis.

Garcia-Villanova et al. [10,11] studied the formation, evolution and modeling of THMs in the drinking water system of a town, both at the treatment facility and in the distribution system. For the first part of this study samples were taken at eight points chosen from two conventional water treatment plants for the city of Salamanca (Spain). The values obtained were correlated statistically with the following parameters: concentration of humic acids, pre- and post chlorination dosages, UV absorbance (UV- 254), pH and temperature. No statistical correlation was observed either with the humic acid content or with the organic matter measured as UV-254. So it was concluded that both pH and temperature increase the concentration of chloroform, although for each pH value all the ln $CHCl_3$ (μg/L) versus temperature curves showed a maximum in correspondence of a temperature value of 18.97°C, after which chloroform levels decrease sharply.

In the second stage of the Garcia-Villanova et al. study [11], six sampling points located at different distances from the three main supply reservoirs were selected in the distribution system of the drinking water network of the city of Salamanca (Spain) in order to follow the evolution of THMs. Data obtained were correlated statistically with the chlorination dosages in the treatment plants and the distribution system, the distances run by the water, the residual free chlorine and total chlorine, the total organic carbon (TOC), pH and temperature. Temperature and pH proved to be the parameters with the strongest influence, where increasing values of pH and temperature are seen to increase the level of $CHCl_3$ up to a given temperature value (T = 17.3°C), after which a sharp decrease in the $CHCl_3$ content occurs.

Nokes et al. [12] developed a simple kinetic model that mathematically relates the extent of bromination and the relative abundance of the four THMs to the [Br^-]:[chlorine] ratio. The results of the study show that chlori-

nation of water containing bromide and natural organic matter (NOM) leads to the formation of brominated THMs. Using US EPA cancer potency factors, the model was used to predict the relative cancer risk associated with THMs as a function of the [Br^-]:[chlorine] ratio. This risk increases steeply to a peak at a [Br^-]:[chlorine] ratio of approximately 0.15, then gradually decreases to the value associated with bromoform alone.

Elshorbagy et al. [13] studied the simulation of THM species in water distribution systems. An approach to characterize and model the kinetics of THM species in a water distribution system was presented. The model is based on nonlinear optimization and capable of modeling chlorine, total THM, and the single four THM species in water distribution systems subjected to different varying loading conditions. The model has been tested and verified by application to a portion of the Abu-Dhabi distribution system in the United Arab Emirates.

The present study focused on the modeling of THMs formation in water distribution systems, so as to develop an improved understanding of the THMs formation process.

2. Materials and methods

In order to simulate the water distribution process, a laboratory-scale unit was constructed. This unit consists of a dual-loop ductile iron pipe network along with a pump, a chlorination unit and a 2 m^3 galvanized steel storage tank. A schematic layout of the network is shown in Fig. 2. The pipes are of 100 mm diameter connected to the tank through a gate valve to control the flow rate. The total length of the pipes in the laboratory network is 42 m.

In order to get data on THM formation and degradation, several experiments were conducted under various combinations of experimental conditions (temperature, chlorine concentration, pH, reaction time). The water used in the experiments was taken from the tap that complies with drinking water quality standards, stored in the tank, where it was subjected to the addition of chlorine and an amount of humic substance was added to reach the required concentration level. The chlorine was added as chlorine gas from a cylinder, while the humic substance was added in the form of humic acid solution prepared from analytical grade from Acros Organics containing 50–60% humic acid. The concentration of humic acid in all experiments was 2.5 mg/L. After the addition of the chemicals, the water was allowed to stay in the reservoir for various periods of time to achieve the necessary reaction time specified in the experimental design (Table 1). The variables examined in the present work were temperature, pH, chlorine dose and reaction time. All variables except chlorine dose were measured after passing the required reaction time. Values of these variables are listed in Table 1. Mixing of water with chemicals was achieved as a result of turbulence that created during tank filling.

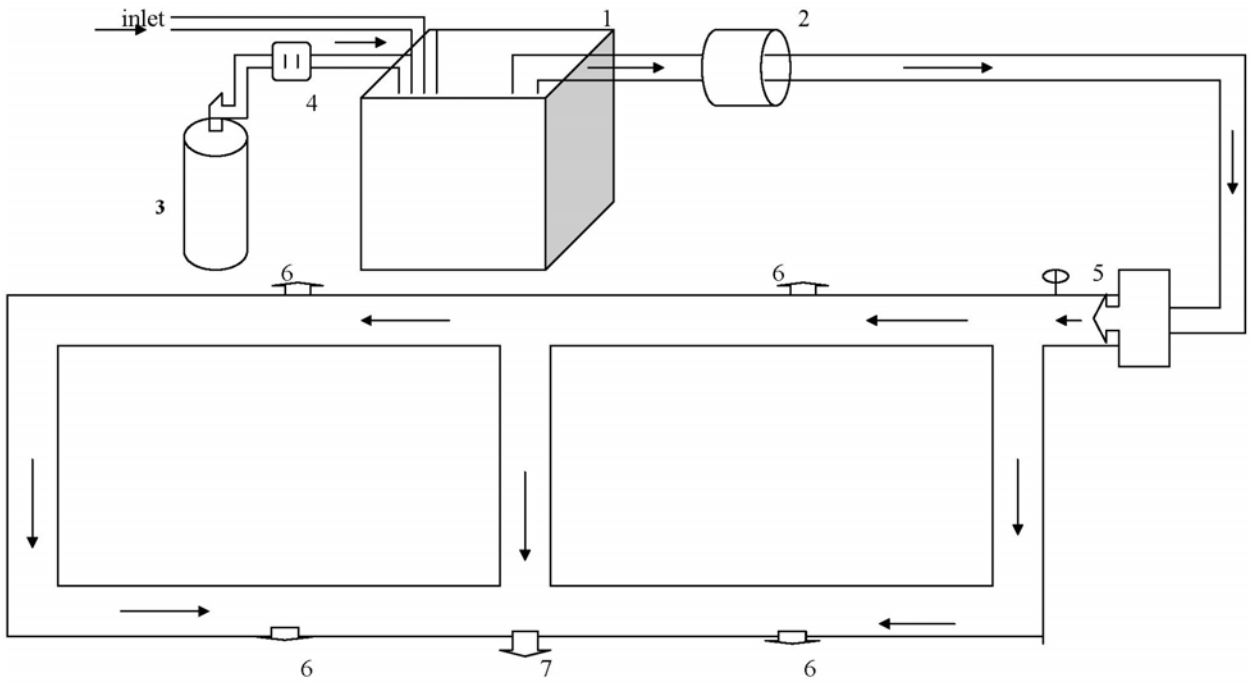

Fig. 2. Schematic diagram of the water distribution network used in the experiments. 1 water tank, 2 water pump, 3 chlorine cylinder, 4 gas flowmeter, 5 control valve, 6 sampling ports, 7 water outlet.

By the end of the contact time, samples of water were withdrawn from the tank. Then, the water was pumped into the network and more samples of water were collected from the sampling ports for THM analysis. The samples were tested for temperature, pH, chlorine and THM measurements. The samples taken from the network for THM analysis were replicate.

2.1. Chlorine concentration measurement

The chlorine concentration was measured by the diethyl-p-phenylenediamine (DPD) method using a Hach DR/820 colorimeter. Both total and free chlorine were measured and the combined chlorine was calculated as the difference between the total and free chlorine values.

2.2. THM measurement

Calibration standard solutions of the four THMs were prepared from pure Supelco compounds and subjected to GC analysis. In the sampling bottles, sodium thiosulfate crystals were added as a preservative to eliminate any residual chlorine and thus stop the THM formation reaction. THM concentrations were determined using head space method [9,14] where the THM in the sample is allowed to volatile in the sampling bottle head space from where it is collected by syringe and injected into a Varian gas chromatograph/mass spectrophotometer (GC/MS-MS) with an MS detector and DB5 capillary column. The carrier gas was helium, and the retention times on the column were about 1.58, 2.036, 2.67, and 3.44 min for chloroform, bromodichloromethane, dibromochloromethane, and bromoform, respectively.

Table 1
Values of the variables that are used in experimental design

Experiment no.	Temp., (°C)	Free chlorine (mg/L)	Reaction time (h)	pH
1	16	21.0	24	7.7
2	19	2.6	24	7.4
3	19	9.6	120	6.8
4	18	9.5	24	7.6
5	17	9.5	48	7.0
6	16	9.5	72	7.5
7	18	9.5	24	7.7
8	17	9.5	48	7.4
9	17	21.0	72	7.3
10	18	21.0	96	7.3
11	17	21.0	24	7.4
12	19	5.7	24	6.7
13	40	21.0	24	7.0

3. Results and discussion

Analysis of 13 samples with replicates showed that THMs were detected in all water samples except two samples. The samples with no THM could be produced as a result of analytical error, as in experiment 4, or from the rapid volatilization of THM due to the high temperature value as in the last experiment, which was carried out at 40°C. The TTHM concentration ranged from 7 ppb to 313 ppb. Table 2 shows the range of variables used in the experiment, their means and standard deviations.

3.1. Effect of temperature

A regression analysis was carried out between lnTTHM and the temperature. As shown in Fig. 3, the relationship is a polynomial of second order with R^2 = 0.7327. From the figure it can be observed that TTHM concentration increases with temperature up to a certain value (i.e., 17°C) after which the concentration decreases. This is may be attributed to the fact that temperature higher than 17°C will lead to THM volatilization. This is also confirmed by the results obtained from the experiments conducted at 40°C, which gave zero TTHM value. A similar conclusion was reached by Garcia-Villanova et al. [11], who noticed that the THM concentration was reduced after the temperature reached about 19°C, which was referred to as a critical temperature.

3.2. Effect of free chlorine concentration

Since the greater THM formation is attributed to free chlorine [11,15], the effect of the free chlorine concentration was considered in this study rather than the total chlorine concentration. Fig. 4 shows the relation between ln TTHM and the free chlorine concentration, which is a second order polynomial with R^2 of 0.795. This suggests that there is a strong correlation between the chlorine concentration and TTHM formation, which is confirmed by the multiple regression, as shown later in the modeling section.

Table 2
Range, mean and standard deviation values of dependent and independent variables

Variable	Min	Max	Mean	Standard deviation
TTHM (ppb)	7	313	126	101.4
Temperature (° C)	16	19	17.58	1.08
Free chlorine (mg/L)	2.6	21	12.45	6.65
pH	6.7	7.6	7.31	0.33
Reaction time (h)	24	120	50	33.09

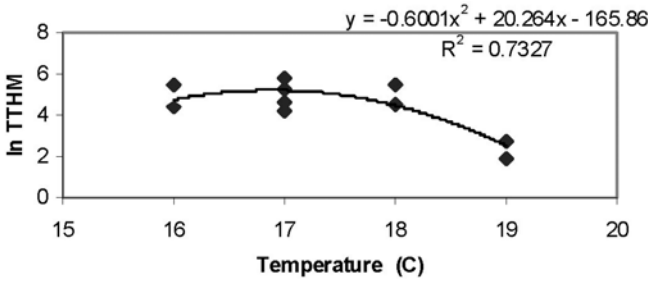

Fig. 3. Relationship between ln TTHM and temperature.

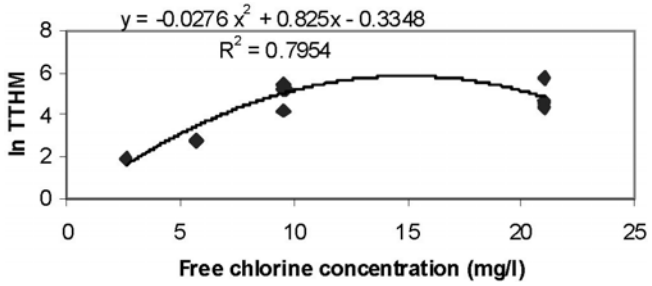

Fig. 4. Relationship between ln TTHM and free chlorine concentration.

3.2. Effect of the reaction time

Several reaction times were considered during the experiments, namely 24, 48, 72, 96 and 120 h (Table 1). Fig. 5 shows the relationship between ln TTHM and the reaction time. As it can be seen from the figure, the relationship between the two variables can be described by a second order polynomial with a relatively low correlation coefficient (R^2 = 0.31). This poor correlation value may be explained by the complex response of THM formation to reaction time. Amy et al. [7] found that the THM formation was overpredicted at longer reaction times, while it was underpridicted at a shorter reaction time. To overcome this problem, a model comprising two submodels was developed: one submodel for short reaction time and the other for long reaction time. Rathbun [15] indicated that TTHM formation as a function of reaction time is a two-phase process: an initial phase where the concentration increased rapidly and a second phase where the concentration increased slowly or was constant.

3.3. Effect of pH

In order to simulate the real characteristics of the drinking water within the distribution systems, the pH values were kept relatively constant (i.e., 6.7–7.7) during the experiments. Thus, within this narrow range of pH values, it is not expected that this parameter will have a significant effect on the THM formation. This will be shown later in the modeling section.

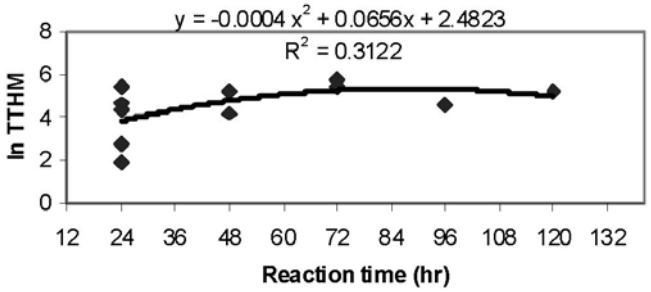

Fig. 5. Relationship between ln TTHM and reaction time.

4. Development of TTHM model

4.1. Modeling approach

Development of TTHM model is the fundamental objective of this study. This can be achieved by developing a suitable prediction equation for the TTHM concentration. As a result, several experiments were conducted under various combinations of experimental conditions (Table 1). The total concentration of TTHM from each experiment was estimated as the sum of the concentrations of the four THM compounds (i.e. chloroform, bromodichloromethane, dibromochloromethane, and bromoform).

Multiple regressions are ones of the most widely used methods in water resources and environmental engineering applications. Regression has been widely used in water quality modeling for predicting THM formation in water distribution systems. Amy et al. [7], Garcia-Villanova et al. [10,11] and Golfinopoulos et al. [16] used the multiple regression techniques to analyze and model the THMs in drinking water systems.

The Statistical Package for Social Sciences (SPSS) software was used to find the best subset regressions for a regression problem. The criterion used to define "best" is R^2 (in percent):

$$R^2 = 100\,[1-(\mathrm{SSE}/\mathrm{SST})] \quad (1)$$

where R^2 is a multiple coefficient of determination which is a measure of the amount of reduction in the variability of TTHM obtained using regressor variables, SSE is defined as the residual sum of squares (residual means the difference between the actual TTHM and the predicted TTHM by the model, and SST is the total corrected sum of squares of TTHM (which is sum of the square of the actual TTHM minus the mean value of TTHM)

4.2. Trihalomethane formation models

One of the most difficult problems in regression analysis is the selection of the model. This is because in most cases the independent variables are not statistically

Table 3
Correlation matrix among the variables used in the regression model

Variable	ln(TTHM)	FCl^2	T^2	pH	T^3	FCl
ln(TTHM)	1.0	0.343	−0.532	0.174	−0.54	0.479
FCl^2	0.343	1.0	−0.461	0.335	−0.467	0.985
T^2	−0.532	−0.461	1.0	−0.575	1.0	−0.528
pH	0.174	0.335	−0.575	1.0	−0.576	0.329
T^3	−0.54	−0.467	1.0	−0.576	1.0	−0.535
FCl	0.479	0.985	−0.528	0.329	−0.535	1.0

independent but are correlated. Therefore, the first step that should be done in regression analysis is to compute the correlation matrix of the independent variables. This correlation matrix is shown in Table 3.

Different types of regression models were investigated using backward elimination for the THM model parameters to select the optimum number of independent variables to be used. The criteria to select the appropriate regression model are a high value of R^2 and the hypothesis that the number of independent variables does not exceed 4. In addition, all the regression coefficients should be significantly different from zero at the five percent level of significance.

In developing the regression model for the TTHM in this study, the values of experimental factors given in Table 1 were used. The independent variables were temperature (T), reaction time (RT), pH, and free chlorine concentration (FCl), while the dependent variable was lnTTHM. Following the approach described in the previous section, the results of the regression analysis was a prediction equation for the TTHM concentration. The SPSS backward regression indicated that there are five possible models as follows:

- Model I:

$$\ln(\text{TTHM}) = -34.35 + 0.598\,FCl - 0.0206\,(FCl)^2 + 0.419\,\text{pH} + 2.86\,T + 7.821\,e^{-5}\,(RT)^2 - 3.311\,e^{-3}\,T^3$$

- Model II:

$$\ln(\text{TTHM}) = -33.849 + 0.566\,FCl - 0.01948\,(FCl)^2 + 3.136\,T + 7.358\,e^{-5}\,(RT)^2 - 3.69\,e^{-3}\,T^3$$

- Model III:

$$\ln(\text{TTHM}) = 1.464 + 0.68\,FCl - 0.02295\,(FCl)^2 + 5.254\,e^{-5}\,(RT)^2 - 2.0\,e^{-4}\,T^3$$

- Model IV:

$$\ln(\text{TTHM}) = -0.235 + 0.787\,FCl - 0.02625\,(FCl)^2 + 2.963\,e^{-5}\,(RT)^2$$

- Model V:

$$\ln(\text{TTHM}) = -0.335 + 0.825\,FCl - 0.02755\,(FCl)^2$$

where FCl is the free chlorine concentration in mg/L, T is the water temperature in °C, and RT is the reaction time in minutes.

The coefficient of determination R^2 and the F-test values of the regression equations are given in Tables 4 and 5, respectively. R^2 ranges from 0.79 to 0.84 for these models. Testing the hypothesis that the regression does not explain a significant amount of the variation in TTHM concentration indicates that this hypothesis is accepted for the first model (I) and rejected for the other four models. As a result, model I is rejected on the basis of the F test. Analyzing the estimated regression coefficients, the standard errors of regression coefficients and the calculated t-value used in testing H_0:$B_i = 0$ (B_i is the regression coefficient) indicates that not all the regression coefficients are significantly different from zero at the 5% level of significance. This is true for all the coefficients in model II. Thus model II is rejected based on the t-test. Model V is the most preferable model among the three remaining models based on both F and t-tests. Model IV is the second preferable model and model III is the least accepted one.

Accordingly, model V, which only considers the free chlorine as an independent variable, explains 80% of the

Table 4
Suggested regression models for the TTHM concentration

Model	R	R^2	Adjusted R^2
I	.917[a]	.841	.602
II	.913[b]	.833	.667
III	.905[c]	.819	.698
IV	.898[d]	.807	.724
V	.892[e]	.795	.744

[a]Predictors (Constant), T^3, RT^2, FCl, pH, FCl^2 ,and T.
[b]Predictors: (Constant), T^3, RT^2, FCl^2, FCl,T.
[c]Predictors: (Constant), T^3, RT^2, FCl^2, FCl.
[d]Predictors: (Constant), RT^2, FCl^2, FCl.
[e]Predictors: (Constant), FCl, FCl^2.

Table 5
Summary statistics of regression results (ANOVA)

Model		Sum of squares	*df*	Mean square	*F*	Significance
I	Regression	11.782	6	1.964	3.523	0.122[a]
	Residual	2.230	4	0.557		
	Total	14.011	10			
II	Regression	11.677	5	2.335	5.003	0.051[b]
	Residual	2.334	5	0.467		
	Total	14.011	10			
III	Regression	11.472	4	2.868	6.775	0.021[c]
	Residual	2.540	6	0.423		
	Total	14.011	10			
IV	Regression	11.302	3	3.767	9.732	0.007[d]
	Residual	2.710	7	0.387		
	Total	14.011	10			
V	Regression	11.145	2	5.573	15.554	0.002[e]
	Residual	2.866	8	0.358		
	Total	14.011	10			

[a]Predictors (Constant), T^3, RT^2, FCl, PH, FCl^2 ,and T.
[b]Predictors: (Constant), T^3, RT^2, FCl^2, FCl, T.
[c]Predictors: (Constant), T^3, RT^2, FCl^2, FCl.
[d]Predictors: (Constant), RT^2, FCl^2, FCl.
[e]Predictors: (Constant), FCl^2, FCL^2.
[f]Dependent Variable, ln(TTHM).

variability in TTHM, while adding the temperature, pH and the reaction time to the model will explain only 4% more in the TTHM variability as shown in Table 4. The strong influence of the free chlorine on the variability of the TTHM, as compared to the other independent variables, is due to the range of the variables values used in the experimental design. In trying to simulate the real water conditions within the distribution networks, the selected range for the pH was 6.8–7.7 and 16–19°C for the temperature (40°C was selected to check the effect on the TTHM volatility), which are close to the real values of the water available in the distribution system. On the other hand, the free chlorine concentration and the reaction time may be within the ranges used in the real water distribution systems, especially when the water is stored for several days in reservoirs as in the case of intermittent water supply.

5. Conclusions and recommendations

Water utilities are faced with several challenges to maintain the water quality in the distribution systems. One of these challenges is the trihalomethanes formation while protecting against microbiological contamination. Water quality modeling is a useful tool in this regard that helps water utilities in establishing operational strategies to comply with regulation requirements.

The present study is an attempt to develop a mathematical model that predicts TTHM formation in the water distribution system at a laboratory scale. A multiple regression technique was used in formulating the model. The independent variables used in the model were temperature, free chlorine concentration, pH and the reaction time. The backward elimination produced five models with different values of correlation coefficients. The most appropriate model to describe the TTHM formation was Model V, which relates the TTHM formation to the free chlorine concentration.

The model developed in this study would serve as a basis to conduct a study on a real water distribution network to enable the water utilities to develop strategies for preventing or minimizing TTHM formation by keeping it below the regulatory levels, while maintaining adequate chlorine residual in the system. It is recommended that further studies are needed to relate the THM formation to pipe network characteristics and conditions.

Acknowledgement

This work was funded by The Higher Council of Science and Technology. The authors are grateful to Eng. Zakarya Al-Tarawneh and Eng. Randa Toffaha from the Water Authority of Jordan for their support during the work on this project.

References

[1] T.A. Bellar, J.J. Lichtenberg and R.C. Korner, J. AWWA, , 66(12) (1974) 703–706.
[2] National Cancer Institute, Carcinogenisis Program, Bethesda, MD, 1976.
[3] US EPA, Criteria and Standards Division, Washington DC, 1990.
[4] R.J. Bull and F.C. Kopfler, Health effects of disinfectants and disinfection by-products, AWWARF and AWWA, Denver, 1991.
[5] V. Uyak, , Environ. Internat., 32 (2006) 12–21.
[6] L.A. Rossman, P.F. Bolous and T. Altman, Planning Management, 119(5) (1993) 505–517.
[7] G.L. Amy, P.A. Chadik and Z.K. Chowdhury, J. AWWA, 79(7) (1987) 89–97.
[8] M.C. Kavanaugh, A.R. Trussell, J. Carmer and R.R. Trussell, J. AWWA, 72(10) (1980) 578–582.
[9] J.C. Lou and P.C. Chiang, Haz. Wastes Haz. Mat., 11(2) (1994) 333–343.
[10] R.J. Garcia-Vilanova, C. Garcia, J.A. Gomez, M. Paz Garcia and R. Ardanuy, Water Res., 31(6) (1997) 1299–1308.
[11] R.J. Garcia-Vilanova, C. Garcia, J.A. Gomez, M. Paz Garcia and R. Ardanuy, Water Res., 31(6) (1997) 1405–1413.
[12] C.J. Nokes, E. Fenton and C.J. Randall, Water Res., 33(17) (1999) 3557–3568.
[13] W.E. Elshorbagy, H. Abu-Qdais and M.K. Elsheamy, Water Res., 34(13) (2000) 3431–3439.
[14] J.W. Li, Z. Yu, M. Gaw and X. Cai, Water, Air Soil Poll., 97 (1997) 367–378.
[15] A.A. Stevens and J.M. Symons, Alternative disinfection processes, in: Organic Carcinogens in Drinking Water, Wiley, New York, 1986, pp. 272–275.
[16] S.K. Golfinopolulos, N.K. Xilourgidis, M.N. Kostopoulou and T.D. Lekkas, Water Res., 32(9) (1998) 2821–2829.