Monitoring and Modeling of Total Trihalomethane Compounds Formation in a Water Treatment Plant in El-Behira Governorate, Egypt.

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ADDITIONAL INDEX WORDS: Chorination, trihalomethanes, water treatment, multiple regression model.

ABSTRACT

A 12 month monitoring program from January 2009 to December 2009 was established for measuring total trihalomethane compounds (TTHMs) and other operational parameters including pH, TOC, chlorine dose and residual, temperature and the bromide ion of both raw and processed water in a water treatment plant in Damanhour, El- Behira Governorate, Egypt. The values of TTHMs measured by GC-ECD analysis ranged from 23±0.44 µg/L to 85 ±1.0µg/L. Values of pH ranged from 7.00±0.04 to 8.26±0.09 and 6.5±0.04 to 7.6 ±0.1 in raw and processed water ,respectively. Ranges of temperature were 18.7 ±0.06°C to 25±0.12 °C and TOC from 3±0.09 mg/L to 30.20±0.9 mg/L found in raw water compared with a temperature range of 20 ±0.1°C to 25 ±0.8°C, and a TOC range of 1.98±0.11 mg/L to 5.0±0.15 mg/L in processed water however Bromide ions were not present in any of the water samples examined. Two multiple linear regression models one for raw water and the other for processed water were established. The R^2 value of the TTHMs model in processed water was 89.9% while 86.9% of raw water and the Durbin - Watson statistic value was 1.772 for processed water compared with 1.549 for raw water suggesting that the model based on processed water parameters is considered more reliable
in predicting levels of TTHMs.

INTRODUCTION

Chlorine is currently the most reliable chemical disinfectant used for effectively killing most micro-organisms. Chlorine is usually added in sufficient quantities that a small residual will be present in treated water through the distribution system. This provides protection to consumers in case bacterial contaminants later gain access to the supply. However during the chlorination of water, a large number of chlorine by-products is formed with more than 300 different types of disinfection by products(DBPs) being identified (Becher,1999). The formation of chlorinated DBPs in drinking water such as trihalomethane(THMs)has emphasized the need for exploring alternative disinfectants and new

treatment technologies. THMs consist of several methane derivative compounds and the four chemical species, known as TTHMs, comprise chloroform (CHCl3), bromodichloromethane (CHBrCl2), dibromochloromethane (CHBr2Cl)and bromoform(CHBr3)(USEPA,2001; Sorlini et al.,2005)These chemicals were classified as possible carcinogens to humans and recent studies suggest a link between adverse reproductive outcomes and exposure to THMs during pregnancy(Dodds *et al.*,1999,King *et al.*,1996)The total concentration of trihalomethanes (TTHMs) and the formation of individual THM species in chlorinated water depend on the composition of the raw water, on operational parameters during water treatment and on residual chlorine in distribution systems (Golfinopoulos,2000).To minimize the risk of cancer, the United States Environmental Protection Agency (USEPA), the World Health Organization (WHO), have introduced regulations for levels of THMs in drinking water(WHO,1993). USEPA and the (WHO) regulate total THM concentrations at 80 µg/L and 100 µg/L respectively. The MCL for THMs compounds in Egypt was set at 100 µg/l in the Egyptian Drinking Water regulations according to the Decision of the Minister of Health No. 458/2007. Several parameters such as pH, temperature, total organic carbon (TOC), bromide ions, residual chlorine, and chlorine dosage and contact time were reported to significantly affect the formation of TTHMs with TOC being the most important factor in the formation of TTHMs (Nikolaou *et al.*,2004).Thus the formation of TTHMs during the processes of water treatment clearly requires monitoring but this is extremely time consuming and involves the use of expensive techniques such as gas chromatography. Alternative approaches are therefore required, so the aims of the present study were to: (a) monitor the TTHMs and other parameters such as pH, temperature, TOC and chlorine in raw and processed water from a treatment plant of EL-Behira Governorate in Egypt and (b) develop predictive models for predicting concentrations of total TTHMs formed during the chlorination of river water.

MATERIALS AND METHODS

Sampling

Water samples were collected from a conventional water treatment plant of Damanhour in El-Behira Governorate,Egypt. Major features of this plant include coagulation, flocculation, sedimentation and filtration, whereas chlorination is the only technique used for disinfection of the water. Pre-

chlorination is applied to the coagulation channels and post- chlorination is applied before water enters the distribution system.
Replicate water samples were taken from January to December 2009 at the entrance to the plant where chlorine is added to raw water and at the exit of the processed water tank. Samples were also taken from the distribution system and measurements of pH, temperature, bromide ions, chlorine and TOC were taken in all samples. Temperature and pH were measured on site, whereas typical chlorine dosage data was collected in the plant, and free residual chlorine of processed samples measured using N,N-diethyl-1,4-phenylenediamine(DPD). For TOC, samples were collected in brown bottles, preserved at pH<2 by adding phosphoric acid and measurements were made with a TOC analyzer (AP0110 -9000). Bromide ion concentrations were measured using a (Dionex-600) ion chromatograph with an electrochemical detector with a column: As –HC (4×250 mm) and analytical conditions of 9 mM -Na2CO3, and a flow rate of 1 ml/min.

Sample bottles for TTHMs measurement were washed with soap and water, rinsed with tap and distilled water. Samples were preserved by the addition of sodium thiosulphate (3mg/40 ml) in screw- capped 40-ml amber glass dark vials to arrest the formation of additional TTHMs and to prevent free chlorine reacting with impurities in the extraction solvent to form organohalides. Sample bottles were half filled, shaked once and then fully filled to prevent air bubbles being trapped in the sealed bottle. All samples were placed in a cooler filled with ice and stored at 4°C until analysed within 14 days of collection (EPA,1996). All methodologies used were described by the standard methods for the examination of water and waste water (21st edition 2005) .

Chemical analysis of TTHMs in water.

Solvents used in this study were pentane- methyl alcohol, acetone, also calibration standards ; (bromoform, bromodichloromethane, dibromochloromethane and chloroform) were of high purity which helped to minimize the interference problems.

1- GC analysis

All samples were analyzed according to the respective method described in the standard methods, (SM6232-B) the liquid –liquid extraction and gas chromatographic method [9]. Samples and standards were allowed to attain room temperature, and after samples extraction they were

injected via autosampler into the gas chromatography. Standards were analyzed and a calibration curve was made.

TTHMs analysis was carried out using a Gas chromatogay (GC – THERMO) with temperature programming equipped with Electron capture detector (ECD) Ni63. A capillary column (RTX-50) 30 ml × internal diameter 0.25µm, 0.5µm film thickness was used. Autosampler was used for injection, (split- mode), with Helium as a carrier gas and Nitrogen as a makeup gas.

2- GC-conditions

The injection and the detector temperature were 220°C and 330°C respectively, Oven temperature was programmed , the initial temperature was 50 °C and was maintained for 1 minute , then increased at 3°C /min up to 180°C and subsequently up to a rate of 10 °C / min up to 220 °C . After standardization, the method blank was analysed (Singer,1994)followed by the samples. To ensure the accuracy of the analysis, calibrations were run after every tenth sample.

3- Model development

A predictive model for the concentration of TTHMs formed during chlorination of water, can be helpful to the minimization of their formation during water treatment, and therefore to compliance with the legislative measures and to protection of human health. The modeling of TTHMs consists of establishing empirical or mechanistic relationships between TTHM levels in processed water and the water quality and water treatment operational control parameters (such as chlorine dose applied, temperature, pH and TOC).

The inclusion of seasonal trends in the formation of THM in the model was deemed necessary, however it was found that there was no general trend for seasonal variation in the formation potentials of TTHMs because there was temporal and spatial variation in the structure of Natural Organic Matter (NOM|) and reactivity of chlorine thus their inclusion in the model was excluded. Furthermore, there was a balance in the flow rate for the different parts of the plant. For that reason the contact time, which is also the chlorine reaction time with the organic substrate, was fairly constant for each step of the treatment plant thus was not included as a variable in the model .

4-Development of multiple linear regression models

For model development and validation, 73 samples were analysed to estimate the concentration of TTHMs in the finished water tank, one based on the characteristic of raw water and the other on those of processed water.

Data Set

For both models the variables included were temperature, pH and total organic carbon content (TOC). The chlorine dose,which was applied to the raw water model, was replaced by residual chlorine in processed water. The development of the model was based on the TTHMs concentration from the finished water reservoir. Both values of prechlorination and postchlorination were used for the chlorine dose , as the reaction continues with the addition of chlorine and contributes to the additional formation of THMs . The Minitab Release 14 software package was used in the model equation and subsequent correlations and the Statistical Package for the Social Sciences (SPSS 10.0) software (SPSS,1999) was used for data analysis. Multiple regression analysis was applied to evaluate statistically significant variables with a level of significanc(α) of 0.05. The inclusion of each variable was based on the t-criterion (Ott ,1988).

Data analysis

Variables were tested for normality using the Kolomogrov- Smirnov (K-S) test, which deals with continious distributions (Conover , 1980)and the goodness-of-fit to normal distributions (Zar, J.1984) .The Pearson correlation coefficient was used to measure the strength of the relationship between variables.

RESULTS AND DISCUSSION.

The results of this monitoring program showed that in water samples collected from the treatment plant, bromide ions were not detected. pH values ranged from 7.00±0.04 to 8.26±0.09 and 6.5±0.04 to 7.6 ±0.1 in raw and processed water, respectively. Temperatures ranged from 18.7±0.06oC to 25±0.21oC and TOC from 3±0.09mg/l to 30.20±0.9 mg/l in raw water compared with a range in temperature of 20 ±0.2oC to 25±0.8 oC, and TOC of 1.98±0.11 mg/l to 5.0±0.15 mg/l in processed water. The maximum operational parameters i.e., pH, temperature and TOC in both raw and processed water were found in the spring and

summer seasons. The concentrations of TTHMs varied significantly and ranged from 23±0.44µg/l to 85±1.0µg/l throughout the year (Fig. 1, 2, 3 and 4). A maximum residual chlorine value of 2.4 ±0.1 mg/l was recorded in the spring compared with maximum values of TTHMs 85±1.0 µg/l and 84±0.4 µg/l in spring and summer respectively. Such an increasing trend during these seasons is due to an increased dose of chlorine for water disinfection, accompanied with higher water temperatures , TOC and pH, compared with lower levels of TTHMs of 77±1.45 µg/l and 61±0.58 µg/l in the autumn and winter respectively. These results are in agreement with those of (Golfinopoulos *et al.*,2002), Uyak *et al.,2005)* and (Rodriguez *et al* . ,2003).

Adin *et al.*,1991.showed that pH demonstrates two effects , with decreasing levels resulting in the low formation of THM and vice versa with an increase in pH. Such differences were due to variation in the concentration of HClO, which increases as levels of pH decrease. Thus when the formation of THM increases, the concentration of HOCl decreases, which in the present case is linked with the residual chlorine. Gracia -Villanova *et al.*,1997 reported that temperature also increases the rate of formation of THMs up to 18.97°C at which the rate of removal of THMs, due to their volatility, exceeds their rate of formation.

Proposed THM models

Two models were developed for predicting the concentration of TTHMs incorporating values of TOC, pH, temperature, with the chlorine dose applied in the raw water model and residual chlorine in the processed water model. The resultant Raw water regression model obtained for the study area was: TTHMs = - 155 + 3.07 CHLORINE DOSE + 0.948 TOC + 3.22 temperature + 12.5 pH.

And for the processed water was: TTHMs = - 230 + 17.7 residual + 23.2 PH + 3.65 TEMPERATURE + 1.16 TOC.Results of K-S tests for the estimating goodness of fit of TTHMs to the models showed that all variables followed a normal distribution at a significance level of p = 0.05 and that all points are close to a straight line, which indicates normality (Adin *et al.*,1991).

1-Raw water model (Table 1.)

The Pearson correlations showed that the temperature has a significant effect on the formation of TTHMs (r=0.780) and that seasonal variations in temperature can influence reaction rates (Uyak *et al.*,2005).Moreover the Pearson correlation matrix also showed a high

correlation (r=0.805) between the formation of THMs and TOC . Previous studies have shown that TTHMs formation increased with a rise in soluble humic material in surface water.It was reported that a large amount of THM production was derived from the humic fraction, which reacts more readily with chlorine than the fulvic fraction. As a consequence of the slow reaction between THM precursors and chlorine, THM formation is a second-order reaction with respect to TOC, especially for long-term THM formation .Thus the formation of THMs involves a multi-stage reaction pathway with an initial fast reaction of chlorine with TOC to produce chlorinated intermediates theses then undergo further slow reactions using several pathways to produce THMs and other by-products (Knocke *et al .,1992)*However the present results showed a moderate relationship between the formation of TTHMs and the chlorine dose (r= 0.570) and also pH (r= 0.598) .

The ANOVA result showed that TOC, temperature, pH and chlorine dose were statistically significant in the formation of TTHMs.

2-Processed water model (Table 2.)

Significant (p=.000) relationships were obtained between THMs formation and residual chlorine (r =0.833) and also pH (r =0.826). The pH of processed water ranged from 6.5±0.04 to 7.6 ±0.17.90, and in this range HOCl is the more prevalent chlorine species which is responsible for the formation of TTHMs , as when the concentration of TTHMs increases, that of HOCl decreases, which in this case is also the residual chlorine (White ,1992). Garcia-Villanova et al.,1997 reported that there was a linear relationship between pH and TTHMs formation, as in the present case, suggesting that pH is important in controlling TTHMs formation. The Pearson correlation matrix also showed a highly significant correlation (r=0.786) between THMs formation and water temperature, but a moderate correlation (r= 0.668) between TTHMs formation and TOC.

The ANOVA results showed that tempertaure, pH , TOC and residual chlorine were statistically significant in the formation of TTHMs.

3-Models validation

The fit of the models was evaluated through the examination of various statistical indicators, including R2 and the F- statistic, while validation focused on the slope and the intercept of predicted vs. measured values (Rodriguez et al.,2003) The model validation requires assessing the effectiveness of the fitted model equation against an

independent set of data ,The number of observations N,F ,R2 ,Standard error of estimate and the Durbin Watson statistic of the proposed models are statistically significant. The values of the Durbin -Watson statistic were found to be 1.549 for Raw water model(Table 3), and 1.772 for the processed water model (Table 4). The value of Durbin – Watson is preferred to be between 1.5 and 2.5 (Nikolaou *et al* .,2004), (Rtledge and Baros 2002), also from Table 4 the processed water model validation indicated satisfactory predictions with R2 value of 0.899 which is higher than that in Raw water model 0.869 Table 3. ,thus the model based on the processed water parameters is considered to be more reliable for the TTHMs prediction .

Statistical evaluation of the models.

1-.Application in the water treatment plant.

Predicted models developed for THMs were validated against individual data sets of TTHMs measurements in water samples taken from the same water treatment plant at a different time –scale in January and February 2010 using identical procedures and chemicals. Under these conditions of variations in pH , chlorine dosage, residual chlorine, TOC and temperature , the formation of TTHMs in water is not stochastic in character, but is more easily described by conventional regression techniques. Modeling results showed that, with the use of these two models , it is possible to estimate TTHMs levels in chlorinated Raw water and processed water in the finished water tank of water treatment plant.

2-Application on field test

The model was tested on one of the water distribution system network of Damanhour city in El- Behira governorate, the values of TTHMs are illustrated in Figure 5. Since the calculated results are close to the measured values in the distribution system, it is suggested that this model is applicable to actual water distribution systems.

CONCLUSION

TTHMs are formed during the chlorination of water containing orgainc matter content.Several parameters such as pH, temperature, total organic carbon (TOC), bromide ions , residual chlorine , chlorine dosage and contact time were reported to be the factors that significantly affect the

formation of TTHMs with TOC being the most important one . The formation of TTHMs during the processes of water treatment clearly requires monitoring but this is extremely time consuming and involves the use of expensive techniques such as gas chromatography , thus a predctive model for the concentration of TTHMs formed during chlorination of water, can be helpful to the minimization of their formation during water treatment. This research resulted in two models that were developed for predicting the concentration of TTHMs in a water treatment plant in El-Behira Governorate ,Egypt.

The number of observations N,F ,R2 ,Standard error of estimate and the Durbin Watson statistc of the proposed models are statistically significant. The values of the Durbin -Watson statist were found to be 1.549 for Raw water model and 1.772 for the processed water model . The processed water model validation indicated satisfactory predictions with R2 value of 0.899 which is higher than that in Raw water model 0.869 ,thus the model based on the processed water parameters is considered to be more reliable for the TTHMs prediction .

The formation of TTHMs in water is not stochastic in character, but is more easily described by conventional regression techniques. Modeling results show that , with the use of these two models , it is possible to estimate TTHMs levels in chlorinated Raw water and processed water in the finished wate tank of that water treatment plant. Future research in this area must be directed to the study of spatial and temporal variations of THMs levels in source water and also modeling studies within the distribution system in El-Behira Governorate , Egypt. However it must be noted thatthe complexity of the TTHMs formation makes it difficult to develop a universally applicable model.

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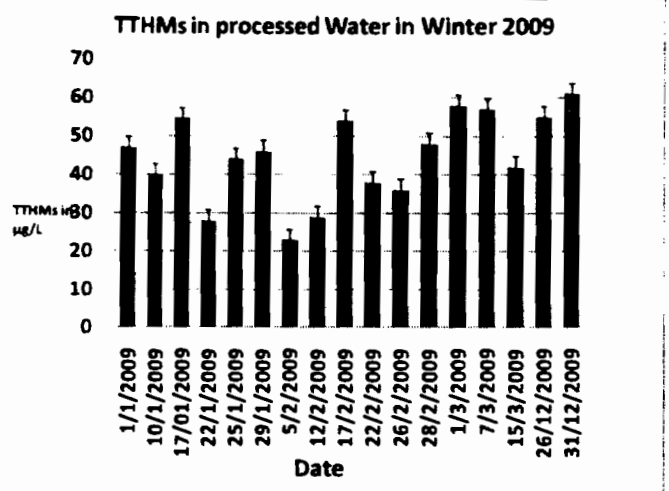

Figure 1: TTHMs in processed water in Winter 2009.

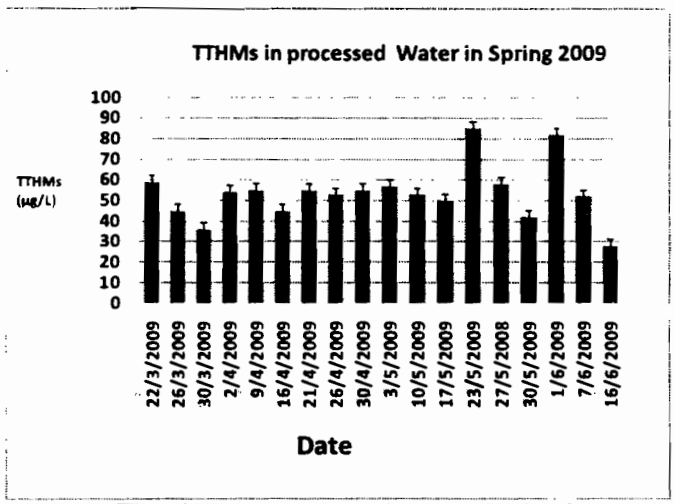

Figure 2: TTHMs in processed water in Spring 2009.

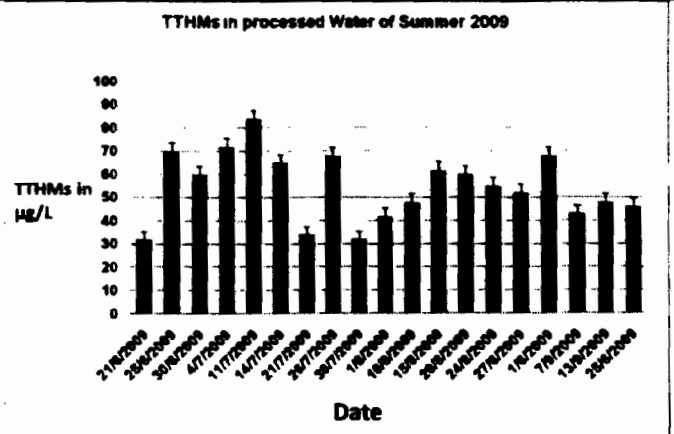

Figure 3:TTHMs in processed water in Summer 2009.

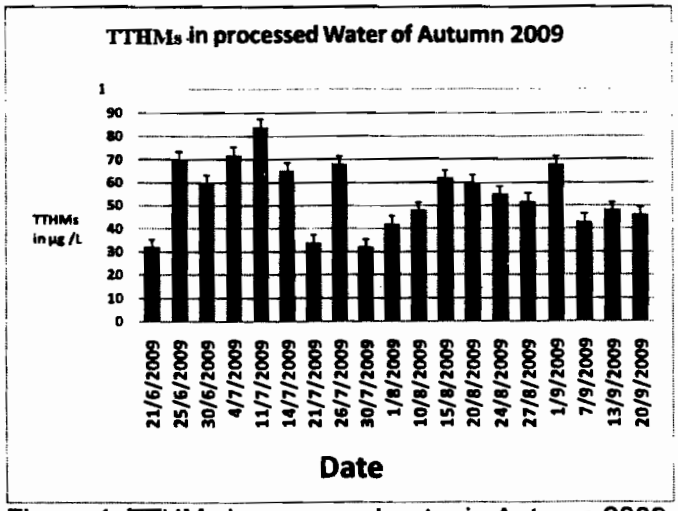

Figure 4: TTHMs in processed water in Autumn 2009.

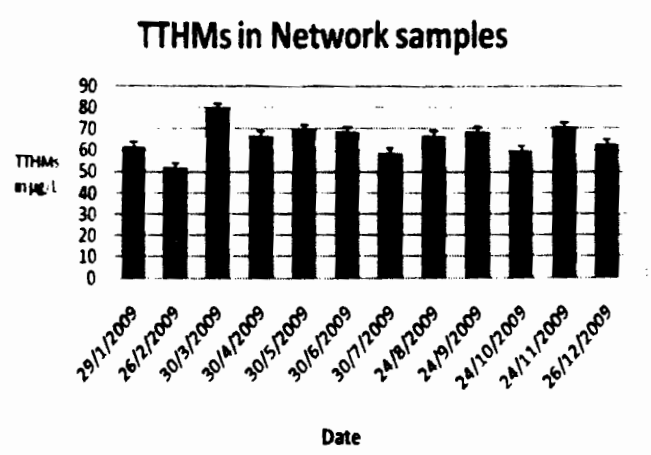

Figure 5: TTHMs in Network Samples

Table 1 : Pearson correlation matrix for raw water quality and operational parameters

		TTHMs	pH	Temperature	TOC	Chlorine Dose
Pearson correlation	TTHMs	1.000	0.598	0.780	0.805	0.570
	pH	0.598	1.000	0.491	0.294	0.397
	Temperature	0.780	0.491	1.000	0.568	0.380
	TOC	0.805	0.294	0.568	1.000	0.464
	Chlorine Dose	0.570	0.397	0.380	0.464	1.000

Sig. (1-tailed) , P=.000

Table 2 : Pearson correlation matrix for processed water quality and operational parameters

		TTHMs	pH	Residual Chlorine	Temperature	TOC
Pearson correlation	TTHMs	1.000	0.826	0.833	0.786	0.668
	pH	0.826	1.000	0.734	0.546	0.578
	Residual Chlorine	0.833	0.734	1.000	0.560	0.671
	Temperature	0.786	0.546	0.560	1.000	0.490
	TOC	0.668	0.578	0.671	0.490	1.000

Sig. (1-tailed) , P=.000

Table 3: Statistical evaluation of raw water TTHMs model.

model	R	R2	Adjusted R2	F value	Standard error of the estimate	Durbin–Watson estimate
Raw water model	0.932	0.869	0.861	112.289	4.95	1.549

Predictors: (Constant), Chlorine DOSE, TEMPERATURE, pH, TOC
Dependent Variable: TTHMs

Table 4: Statistical evaluation of processed water TTHMs model.

model	R	R2	Adjusted R2	F value	Standard error of the estimate	Durbin–Watson estimate
Processed water model	0.948	0.899	0.893	151.05	4.34	1.772

Predictors: (Constant), TOC, TEMPERATURE , pH, RESIDUAL chlorine
Dependent Variable: TTHMs

الملخص العربى

رصد و نمذجة إحصائية لتكوين المركبات الكلية للترايهالوميثان فى محطة لمعالجة مياه الشرب بمحافظة البحيرة بجمهورية مصر العربية

هشام زكى ابراهيم – ولاء عصران

معهد الدراسات العليا والبحوث – جامعة الإسكندرية

قسم الدراسات البيئية

الإسكندرية – جمهورية مصر العربية

مركبات الترايهالوميثان العضوية الكلية هى أحد أشهر نواتج تعقيم المياه بالكلور والتى ثبتت خطورتها على الصحة لكونها من مسببات السرطان ، لذا تم عمل برنامج لرصد تكونها فى الفترة من يناير 2009 حتى ديسمبر2009 ، كما تم رصد بعض ظروف التشغيل الأخرى والمرتبطة بعملية معالجة المياه مثل رقم الأس الهيدروجينى ، الكربون العضوى الكلى ، جرعة الكلور والكلور المتبقى ، درجة الحرارة وتركيز أيون البروم فى كل من المياه قبل المعالجة والمياه بعد المعالجة فى محطة معالجة لمياه الشرب فى دمنهور بمحافظة البحيرة بجمهورية مصر العربية . تم قياس مركبات الترايهالوميثان الكلية بواسطة جهاز الكروماتوجرافى الغازى ملحق به (ECD) وكانت قيمتها تتراوح من 23 ± 0.44 إلى 85 ± 1 ميكروجرام / لتر ، رقم الأس الهيدروجينى يتراوح من 7 ± 0.04 إلى 8.26 ± 0.09 و 6.5 ± 0.04 إلى 7.6 ± 0.1 فى كل من المياه قبل المعالجة والمياه بعد المعالجة على التوالى وكانت درجة الحرارة تتراوح من 18.7 ± 0.06 رقم سيليزية إلى 25 ± 0.12 درجة سيليزية والكربون العضوى الكلى يتراوح من 3 ± 0.09 مجم/لتر إلى 30.2 ± 0.9 مجم/لتر فى المياه قبل المعالجة, وقد كانت درجة الحرارة فى المياه المعالجة تتراوح من 20 ± 0.1 رقم سيليزية إلى 25 ± 0.8 درجة سيليزية والكربون العضوى الكلى من 1.98 ± 0.11مجم/لتر إلى 5 ± 0.15 مجم/لتر بينما تلاحظ عدم وجود أيون البروم فى كل من المياه قبل المعالجة والمياه بعد المعالجة.

ولكون عملية تكون مركبات الترايهالوميثان مكلفة وتستهلك الكثير من الوقت لذا تم عمل نموذجين خطيين متعددى العوامل: الأول خاص بالمياه قبل المعالجة والثانى خاص بالمياه بعد المعالجة . وكانت قيمة R^2 للنموذج الأول = 86.9% وللنموذج الثانى 89.9%. كما تبين من قيمة الاختبار الإحصائى دربين واتسون والتى كانت 1.549 للنموذج الخاص بالمياه قبل المعالجة تقابلها 1.772 للمياه المعالجة أن النموذج الذى تم إنشاءه معتمدًا على قيم المياه بعد المعالجة هو الأفضل للتنبؤ بتركيز الترايهالوميثان العضوية الكلية .