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Modeling of trihalomethanes in the water distribution network of Brazzaville, Congo

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Abstract

The preparation of clean water from a hygienic point of view is one of the main challenges of our time. It is therefore essential to carry out an appropriate treatment of water, in order to avoid any risk to health. In this study, THMTs were identified and quantified in the drinking water distribution network during two sampling periods, namely September 2016 and March 2017, which represent the dry and rainy seasons respectively, with other parameters such as total organic carbon (TOC), temperature (T), pH, concentration of consumed chlorine (DCl), absorbance (UV 254nm), residual chlorine concentration (Cl_2 res.), bromide ion (Br-) and ammonium ion (NH_4^+). THMT are mainly composed of chloroform, which accounts for 97% of THMT. The parameters for better prediction of THMT concentration were: TOC, T and DCI with a coefficient of determination $R^2 = 0.92$.

Keywords: Surface water, chlorination, trihalomethanes, linear multiple regression.

Introduction

The preparation of clean water from a hygienic point of view is one of the main challenges of our time. Water is vital in the life of every living being, thus the well-being of man is also associated with a good quality of drinking water[1]. Disinfection is an important step in the modern treatment of water; the chlorine is used more and more. It is a very effective method for controlling waterborne diseases caused by pathogenic micro-organisms[2]. The network under study is fed by two factories of the National Society of Water Distribution (SNDE) that treat raw water taken from two different rivers (DJOUE and DJIRI). Chlorination is the only disinfection treatment in place in both plants. The DJIRI plant performs pre-chlorination, pH adjustment with lime prior to coagulation-flocculation.

It is therefore essential to carry out a suitable treatment of these waters, in order to comply on the one hand with drinking water standards and, on the other hand, to avoid any health risks related to any toxicity. The latter may be due to substances in the water, or to chlorination by-products formed after treatment[3].

When the water is pumped from the points of capture, they undergo coagulation-flocculation after the addition of aluminum sulphate. The water is then decanted and filtered with sand filters to remove impurities. A final stage of chlorine disinfection is performed prior to storage and distribution of water. By reaction with organic matter and bromides contained in water, chlorine leads to the formation of chlorination by-products, the main ones being chlorinated and brominated as trihalomethanes (THMT), as well as organohalogen compounds such as (COX)[4,5]. The concentrations of chlorination by-products measured in water are related to the intensity of the physicochemical reactions conditioned, in particular, by the chlorine dose, the pH, the contact time, the temperature, the natural organic matter (NOM), bromide ions and environmental conditions (seasonal variability)[6]. Total organic carbon (TOC) and ultraviolet absorbance at a specific wavelength of 254 nm (UV-254) are usually taking in account in the determination of THMT[7].

However, over the past forty years, the development of analytical techniques has made it possible to identify a large number of organohalogen compounds as by-products of chlorination, especially trihalomethanes, which may have a chronic toxicity such as mutagenic and carcinogenic effects[8-10]. The main THMT formed are chloroform ($CHCl_3$), which represents the highest proportion of this family, Bromodichloromethane ($CHBrCl_2$), dibromochloromethane ($CHBr_2Cl$) and bromoform ($CHBr_3$)[11]. The presence of chlorination by-products in drinking water systems justified the setting of regulatory limits.

In several countries, water regulators have imposed maximum levels of maximum limits for drinking water. For example, the United States Environmental Protection Agency (USEPA) prescribed SPC levels in stages I and II. In Stage I the maximum concentration level of THMT was set at 80 µg/L, while in stage II the maximum concentration level of THMT required was

40µg/L to reduce the level of potential risk to human health[12-14]. Similarly, Canada and Australia-New Zealand set the maximum concentration of THMT at 100µg/L and 250µg/L respectively, while in Italy a more restrictive regulation set the limit value at 30µg/L[15,16]. Taking into account the risk to health, a better understanding of their presence and their evolution through the distribution network is necessary[17]. Currently, there is a lack of information regarding THMT levels in the Brazzaville water distribution system. This study aims to identify and quantify the THMT formed and likely to coexist in the water distribution network of the SNDE in Brazzaville, and then develop a mathematical model to predict the concentrations of THMT formed according to operational parameters and water quality.

Materials and methods

Location of the study area: Located on the right bank of the Congo River, Brazzaville is the capital of Congo. It has nine districts and its geographical position is 4°16'04" South Latitude and 15°16'31" East Longitude. In this city are located two water treatment plants, the Djoué plant in south of the city located in the district 1 Makélékélé (Figure-1) and the Djiri plant in north of the city located in the district 9 Djiri (Figure-2). Each of the two plants processes 122,400m^3/day and 163,200m^3/day, respectively.

Sampling: Each sampling point is characterized by geographic coordinates using a Garmin E-Trex GPS (Table-1, Table-2). The sampling covered two seasons: dry season (2016, September) and rainy season (2017, March). The samples were taken at ten points of the network and each point was located at distant 1.5 km from the other (Figure-2, Figure-3). Bottles of 1000 ml glasses were used. Before use, these vials were thoroughly washed and rinsed. They were then dried and then wrapped in aluminum foil and sterilized by autoclaving (125°C) for 15 minutes.

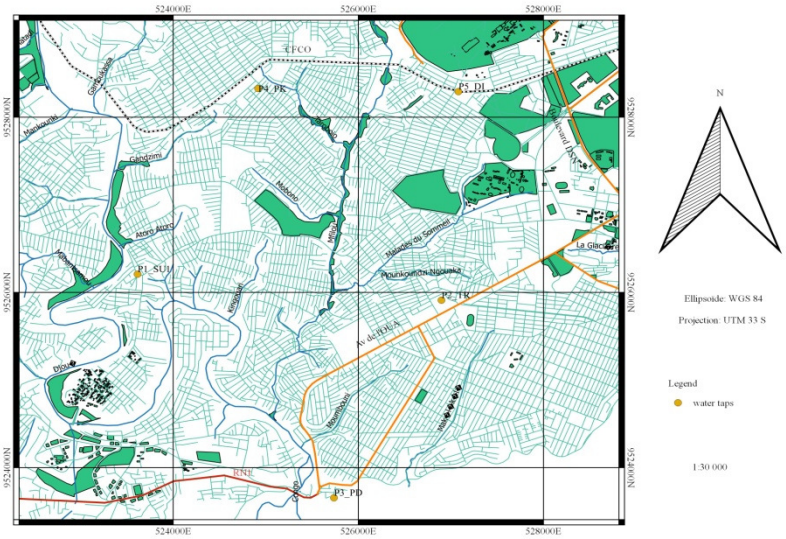

Figure-1: Location of water taps in south of Brazzaville.

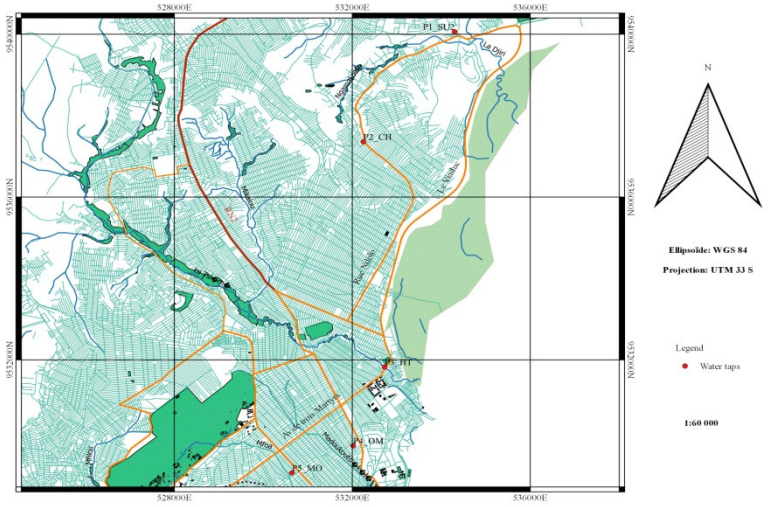

Figure-2: Location of water taps in north of Brazzaville.

Table-1: Geographical coordinates of tap water samples (South Area).

Sampling point	X	Y
P1_SU1 (Plant outlet Djoué)	523614.67	9526208
P2_TR (Terinkyo)	526898.38	9525910.6
P3_PD (Djoué bridge)	525736.96	9523654.9
P4_PK (PK Station)	524918	9528327.3
P5_DI (District Diata)	527081.33	9528292

X: Latitude, Y: Longitude

Table-2: Geographic coordinates of tap water samples (North Area).

Sampling point	X	Y
P1_SU2 (Plant outlet Djiri)	534298.94	9540060.5
P2_CH (Ngamakosso water tower)	532246.46	9537357.6
P3_HT (Talangai hospital)	532724.44	9531825.2
P4_OM (District Ouenze mazanza)	532019	9529891.3
P5_MO (District Moungali)	530641.33	9529226.7

Analytical methods: The following parameters were determined in the water samples taken from the Brazzaville water distribution network: temperature (T), pH, turbidity (Turb.), residual chlorine (Cl_2 res.), total organic carbon (TOC), ammonium ion (NH_4^+), bromide ion (Br^-), absorbance 254 nm (Abs. UV) and THMT. The temperature was measured using a WTW brand thermometer, EU, HACH. The pH was measured using a pH-meter AR25 type, Fischer scientific. Turbidity was measured using an HACH Lange Nephelometric Infrared Light Turbidimeter (Model 2100P IS, Range 0.001 to 1000NTU, Noisy Le Grand, France). Residual chlorine was determined using the DPD method (diethyl-p-phenyldiamine) using a TestpaK apparatus (Comparator 2000+ Lovibond, Tintometer-Group, France). The bromide ion was determined using SpectroDirect / PC spectro II-3 O4/2008 using the pellet method (DPD n°1). Ammonium (NH_4^+) ions were measured using a SpectroDirect / PC spectro II-3 O4 / 2008 using the 2-3 indophenol method. The TOC contents were determined using a SpectroDirect / PC spectro II-3 O4 / 2008 brand analyzer using the MERCK Spectroquant (R) method, cuvette test No. 1.14879.0001, Persulfate method. UV absorbance was measured at 254 nm using SpectroDirect / PC spectro II-3.

THMT analysis methods: The THMT analyzes were carried out by the Laboratory "Environmental Laboratory Services" of the « Société Générale de Surveillance» (SGS France) via SGS CONGO. This laboratory has Cofrac accreditation (French Accreditation Committee). Sample collection for THMT analyzes is done using 40mL glass vials for auto-sampler. The elimination of residual free chlorine is done by adding 4 mg of sodium thiosulfate per vial of 40mL. The method used[18] refers to DIN EN ISO 10301.

The analysis technique includes a concentration phase of THMT by dynamic headspace (Purge and Trap), coupled with an Agilent brand gas chromatograph (model 6890N, Santa Clara, CA, USA), equipped with an Agilent mass spectrometry detector (model 5973, Santa Clara, CA, USA). The limit of detection was 0.1µg/L for most of the compounds measured. The association between THMT and other parameters was highlighted using the binary diagrams. Prediction of THMT concentration was determined using multiple linear regression[19].

This is described as:

$$y = Cte + a.x + b.z + ... + n.m \quad (1)$$

Where: x, z …m are the descriptors, and y the dependant variable.

Except T (°C), pH (unit), Turb. (NTU unit), Abs. UV (cm^{-1}), THMT (µg/L), the others parameters are expressed in mg/L.

Results and discussion

Physico-chemical parameters of tap water samples: The values of the different parameters in the tap water samples for the two areas are given in Table-3, Table-4 for the dry season and in Table-5, Table-6 for the rainy season. The results distinguish the distribution of parameters at the exit of the plants and the distribution network, the effect of pre-chlorination and a description of the spatial and seasonal variations between the outlet and the distribution network. Reading Tables-3, 4, 5 and 6 highlights the effect of treatment channels on the formation of THMT. The die with pre-chlorination (Djiri plant) differs from the chlorine-only sector (Djoué plant) with higher THMT levels regardless of the sampling point and the season. This observation shows that the amount of THMT formed is all the more sensitive to the use of chlorine. The measurements revealed an increase in the THMT concentrations between the output of the plants and the distribution network (spatial variability) and the two higher values P5_DI (40.5 µg/L) and P5_MO (40µg/L) are obtained during the measurements of the dry season (seasonal variability)[20]. Several limiting values of THMT concentrations are encountered in the literature. Thus, according to the restrictive guideline values of Italy and the USA, which respectively limit the maximum concentrations of THMT to 30µg/L and 40µg/L, we can see that three points in the network pose a real health problem, points P5_DI (40.5 µg/L), P5_MO (40µg/L) in the dry season and P3_HT (36.5 µg/L) in the rainy season). These points deserve regular oversight measures to monitor the evolution of the THMT level. However, the study network does not pose a health risk for Canada, Australia-New Zealand and World Health Organization (WHO) guideline values[21-22].

Indicators of organic matter (TOC, UV Abs) were measured between the output of the plants and the different sampling points in the network. The results show a reduction in TOC along the distribution network in both seasons. The highest TOC values were observed in rainy season. The decrease of the TOC values in the water distribution network is accompanied by that of the residual chlorine as described by the correlation coefficient between the two variables (r=0.52) (Table-9). This observation therefore reflects the reactivity of chlorine on organic matter[23]. Other parameters change little in the network.

Table-3: Values of tap water parameters in the southern zone (dry season).

Points	T	Turb.	pH	$Cl_{2\ res.}$	NH_4^+	Br^-	Abs. UV	TOC	THMT
P1_SU1	24.3	5.4	6.5	0.2	0.01	0.29	0.081	7.8	9.4
P2_TR	24.8	6.95	7.36	0.1	0.01	0.44	0.067	6.5	12.2
P3_PD	23.7	3.28	7.41	0.05	0.8	0.27	0.045	3.7	11.5
P4_PK	24.1	2.52	7.41	0	0.09	0.48	0.053	4.2	10.6
P5_DI	25.3	7.46	7.67	0	0.01	0.68	0.041	3.3	40.5
Mean	24.44	5.122	7.27	0.07	0.184	0.432	0.0574	5.1	16.84
S.D	0.48	1.77	0.30	0.06	0.24	0.12	0.01	1.64	9.46

Table-4: Values of tap water parameters in the northern zone (dry season).

Points	T	Turb.	pH	$Cl_{2\ res.}$	NH_4^+	Br^-	Abs. UV	TOC	THMT
P1_SU2	24.2	1.3	6.5	1.21	0.28	0.86	0.071	6.3	11.6
P2_CH	23.7	1.6	6.35	0.5	0.26	0.56	0.069	5.9	19.3
P3_HT	24.3	0.3	7.22	0.4	0.19	0.73	0.055	4.1	22.7
P4_OM	24.1	0.29	7.26	0	0.26	0.46	0.03	2.8	22.1
P5_MO	24.7	0.41	7.83	0.2	0.32	0.64	0.005	1.1	40
Mean	24.2	0.78	7.032	0.462	0.262	0.65	0.046	4.04	23.14
S.D	0.24	0.53	0.48	0.31	0.03	0.11	0.02	1.67	6.74

Table-5: Values of Tap Water Parameters in the southern zone (rainy season).

Points	T	Turb.	pH	$Cl_{2\ res.}$	Br^-	Abs. UV	TOC	NH_4^+	THMT
P1_SU1	22.9	7.9	6.8	1.5	0.04	0.202	17.3	0.09	13.5
P2_TR	23.4	6	6.6	0.2	0.06	0.198	15.5	0.07	15.5
P3_PD	22.8	0.4	7.2	0.1	0.05	0.095	12.2	0.05	7.8
P4_PK	23.7	0.9	6.8	0.3	0.07	0.056	7.3	0.0015	27.5
P5_DI	23.3	1.2	7.1	0.3	0.05	0.07	8.8	0.0017	20.5
Mean	23.22	2.92	6.90	0.48	0.054	0.12	12.22	0.0426	16.96
S.D	0.29	2.08	0.20	0.40	0.008	0.06	3.34	0.03	5.63

The compilation of the results obtained on the various sampling points in the two seasons made it possible to give an average characterization of the water taken from each sampling point of the distribution network. Table-7 and Table-8 give the average values of the water parameters in the two study areas, Djoué plant and Djiri plant, respectively. These average values show that the points P5_DI (30.5µg/L) and P5_MO (30,75µg/L) should be checked regularly in the context of the restrictive limit of THMT in drinking water (30µg/L)[16]. The parameter DCl represents the difference in chlorine concentration between the plant outlet and any sampling point.

THMT modeling: Correlations between THMT and other parameters: Previous research has shown the main factors that affect THMT formation such as residual chlorine dose, consumption of chlorine, total organic carbon concentration, contact time, pH and temperature[24.25]. For this purpose, the average values of the parameters in the ten sampling points of the two study areas were used in the calculation of the correlation matrix.

The existence of possible associations between the physicochemical parameters analyzed was evidenced from correlation matrix taking into account the ten samples (Table-9). Ammonium ions were not taken into account in the calculation of the correlation matrix because they are not involved in the formation of THMT. Table-9 shows the correlation between the parameters.

Table-6: Values of tap water parameters in the northern zone (rainy season).

Points	T	Turb.	pH	$Cl_{2\ res.}$	Br^-	Abs. UV	TOC	NH_4^+	THMT
P1_SU2	23.5	2.7	7.6	0.4	0.06	0.109	16.6	0.08	24.5
P2_CH	23.7	3.1	7.1	0.3	0.07	0.074	9.7	0.026	28.5
P3_HT	24.1	2.3	7.1	0.1	0.09	0.086	10.4	0.06	36.5
P4_OM	23.6	2.2	7.2	0.1	0.08	0.041	6.6	0.003	28.5
P5_MO	23.2	3.4	7.2	0.2	0.09	0.032	5.5	0.0028	21.5
Mean	23.62	2.74	7.24	0.22	0.078	0.068	9.76	0.0343	27.9
S.D	0.22	0.4	0.14	0.10	0.01	0.02	2.99	0.028	3.92

Table-7: Average values of water parameters in the southern area (Dry and rainy seasons, Djoué plant).

Points	T	pH	TOC	Abs.	$Cl_{2rés.}$	DCl	Br^-	NH_4^+	THMT
P1_SU1	23.6	6.65	12.55	0.141	0.85	0.9	0.165	0.05	11.45
P2_TR	24.1	6.98	11	0.132	0.15	1.6	0.25	0.04	13.85
P3_PD	23.25	7.3	7.95	0.07	0.075	1.67	0.16	0.425	9.65
P4_PK	23.9	7.1	5.75	0.054	0.15	1.6	0.275	0.045	19.05
P5_DI	24.3	7.38	6.05	0.055	0.15	1.6	0.365	0.005	30.5

Table-8: Average values of water parameters in the northern area (Dry and rainy seasons, Djiri plant).

Points	T	Turb.	pH	TOC	Abs.	$Cl_{2res.}$	DCl	Br-	NH_4^+	THMT
P1_SU2	23.85	2	7.05	11.45	0.09	0.8	0.7	0.46	0.18	18.05
P2_CH	23.7	2.35	6.72	7.8	0.071	0.4	1.1	0.315	0.143	23.9
P3_HT	24.2	1.3	7.16	7.25	0.07	0.25	1.25	0.41	0.125	29.6
P4_OM	23.85	1.24	7.23	4.7	0.036	0.05	1.45	0.27	0.131	25.3
P5_MO	23.95	1.9	7.51	3.3	0.019	0.2	1.3	0.37	0.161	30.75

There was a low correlation between THMT and T (r=0.48), a medium correlation between THMT and TOC (r=0.506) and no correlation between the consumed chlorine (DCl) and THMT (r = 0.002).

Simple linear regression: The relationship between THMT – TOC, THMT – T and THMT – DCl are shown in Figures-3, 4 and 5, respectively.

The relationships between the THMT and TOC, DCl are not satisfactory because of the low values of R^2, such as 0.04 and 0.68, respectively. The obvious lack of simple relations between the THMT and each of the three descriptors demonstrates that the formation of THMT doesn't depend only on one parameter, but also on the other physico-chemical parameters of the water in the network.

Linear Multiple regression: Three descriptors were retained namely TOC, T and DCl. These three explanatory descriptors selected were determined by mathematical regressions according to the "Back Ward" method[26]. The three descriptors were integrated for the statistical development of the predictive model of THMT training. Correlations between these descriptors and the THMT are shown in Table-10.

Table-9: Correlation matrix of the parameters.

Parameter	T	pH	TOC	Abs. UV	DCl	Cl_2 res.	Br^-	THMT
T	1							
Ph	0.08	1						
TOC	0.06	0.53	1					
Abs. UV	0.02	0.56	0.90	1				
DCl	0.02	0.27	0.26	0.10	1			
Cl_2 res.	0,04	0.41	0.52	0.33	0.85	1		
Br^-	0.41	0.09	0.05	0.14	0.14	0.02	1	
THMT	0.48	0.24	0.51	0.49	0.002	0.10	0.49	1

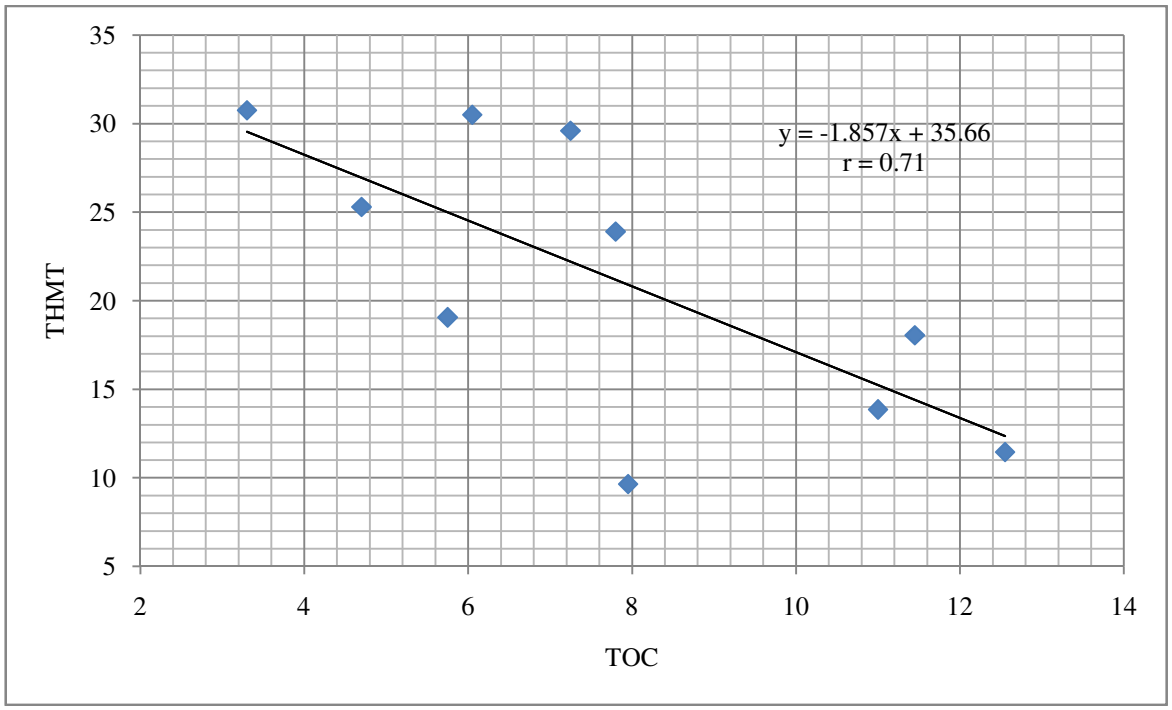

Figure-3: Relation between THMT and TOC.

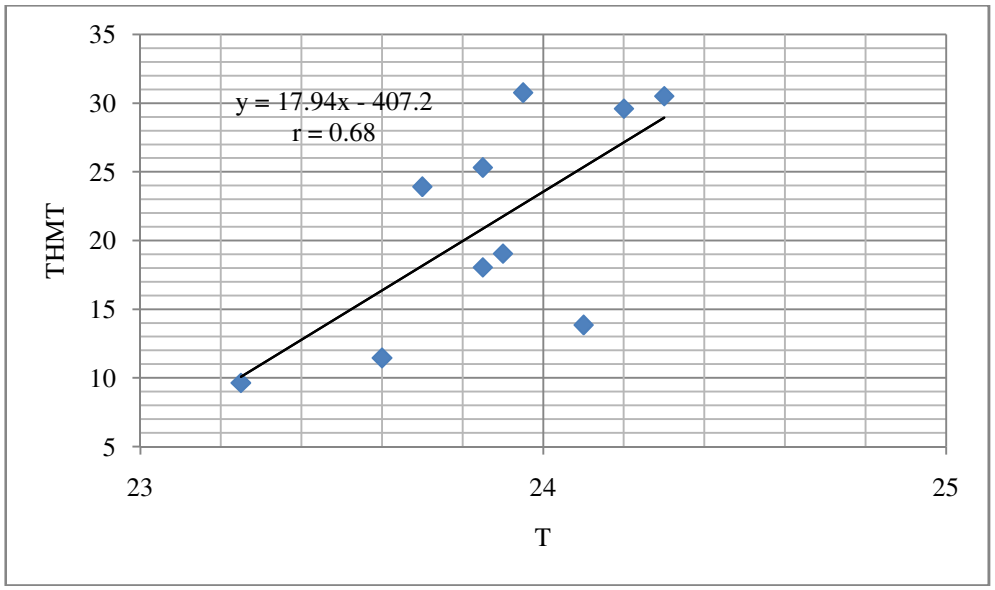

Figure-4: Relation between THMT and T (° C).

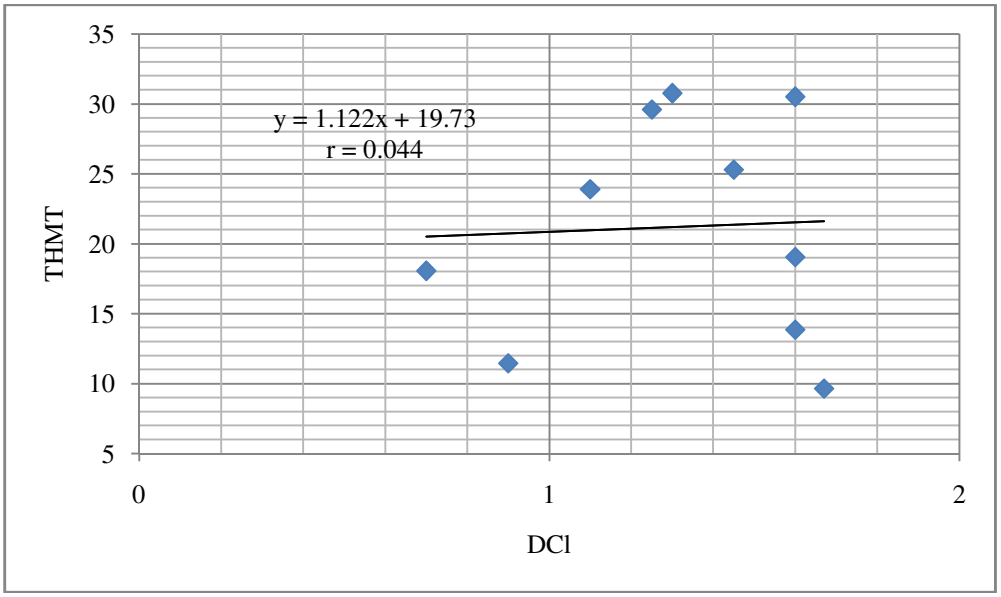

Figure-5: Relation between THMT and chlorine consumption (DCl).

The three descriptors are integrated in equation (1) which represents the model equation. By this model, equation (2) describes the concentration of THMT as a function of three descriptive variables with a correlation coefficient $R^2 = 0.9292$ at $P < 0.05$, which means that they are all significant in the regression model.

THMT = - 291.43 + 14.3466(T) - 2.0867 (TOC) - 10.3122 (DCl) (2)

From equation (2), predicted THMT values were determined and are shown in Table-11. It appears that the values calculated from the model are relatively close to those measured. The evolution of THMT in the distribution network seems to be well represented by the combination of the three descriptors selected.

The distribution of measured and calculated THMT concentration values is shown in Figure-6. This shows the performance of the resulting model. In terms of probability, the TOC parameter is the main descriptor that explains the formation of THMT with a probability (Prob>|T|) of 0.000821 (Table-10)[27-28].

Indeed, the probability (Prob>|T|) reflects the contribution of a parameter to the linear regression. The lower the probability (Prob>|T|) of the parameter, the more significant is its contribution.

Table-10: Linear multiple regression between THMT and T, TOC, DCl.

Analysis of the variance					
Source of variation	Degree of freedom	Sum of squares	Mean square	Value F observed	Prob. > F(p)
T	1	270.956	270.956	40.313	0.0007154
TOC	1	180.424	180.424	26.843	0.0020522
DCl	1	77.676	77.676	11.556	0.0145051
Residue	6	40.328	6.721		

Coefficient of determination
$R^2 = 0.9292$

Parameter estimation				
Variable	Parameter	Typical error of the estimator	T for Ho	Prob (>|T|) p
T	14.3466	2.9116	4.927	0.002637
TOC	-2.0867	0.3373	-6.187	0.000821
DCl	-10.3122	3.0335	-3.399	0.014505
Constant	-291.4275	70.2806	-4.147	0.006034

Table-11: Comparison between measured and calculated THMT concentrations.

Points	T (°C)	TOC (mg/L)	DCl (mg/L)	THMT measured (µg/L)	THMT calculated (µg/L)
P1_SU1	23.6	12.55	0.9	11.45	11.72
P2_TR	24.1	11	1.6	13.85	14.91
P3_PD	23.25	7.95	1.67	9.65	8.37
P4_PK	23.9	5.75	1.6	19.05	23.02
P5_DI	24.3	6.05	1.6	30.5	28.13
P1_SU2	23.85	11.45	0.7	18.05	19.67
P2_CH	23.7	7.8	1.1	23.9	21.02
P3_HT	24.2	7.25	1.25	29.6	27.8
P4_OM	23.85	4.7	1.45	25.3	26.04
P5_MO	23.95	3.3	1.3	30.75	31.95

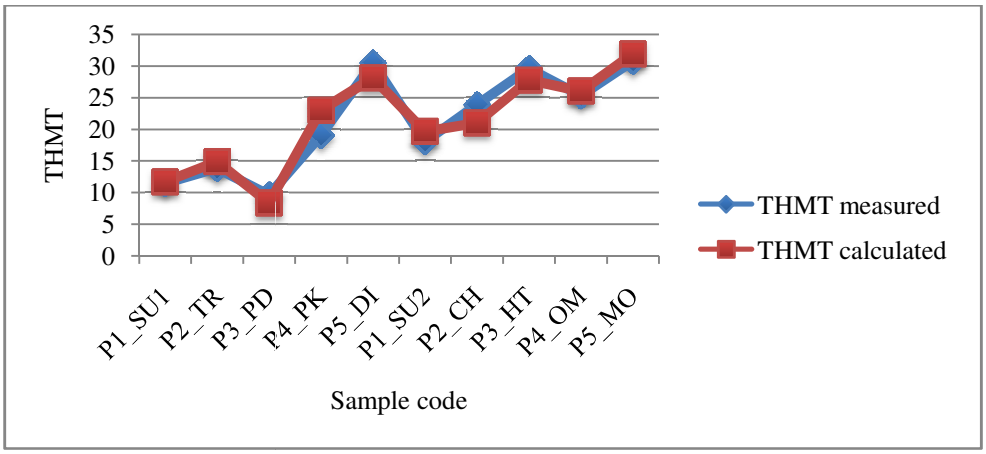

Figure-6: Evolution of predicted and observed THMT concentrations.

Conclusion

In this study, the concentration of THMT present in the water distribution network in Brazzaville was determined. A mathematical model predict the concentration of THMT formed in the different points of the distribution network according to the operational parameters and the quality of the water. It confirmed the importance of experimental parameters such as T, DCl and TOC in the formation of THMT. Taking into account the studied variables, the model appears usable to predict levels of THMT at any point of the network fed from the two rivers. For this, a mathematical prediction equation has been proposed. The performance of the predictive model equation is satisfactory (R^2 = 0.92 close to 1).

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