Journal of Chemical, Biological and Physical Sciences

An International Peer Review E-3 Journal of Sciences

Available online at www.jcbsc.org

Section A: Chemical Science

CODEN (USA): JCBPAT

Research Article

Trihalomethanes in Tetova's Drinking Water

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Received: 15 January 2013; **Revised:** 22 January 2013; **Accepted:** 24 January 2013

Abstract: The formation of carcinogenic trihalomethanes (THMs) in the process of water disinfection by chlorine has raised concerns in the scientific community as well as in the public opinion. This study aims to determine the concentration of THMs in Tetova's drinking water during the summer season and compare it with the regulation in the Republic of Macedonia, the European Union, and the World Health Organization. To this end, we have used the UV-VIS spectrophotometric method based on Fujiwara's reaction. The THMs concentration measured in fifteen different locations in June, July and August 2011. The results indicate that the summer variation is below the critical values stipulated in the state, EU, and WHO regulations (seasonal average 24.75 ± 12.33 µg/L). This study is the first of its kind on THMs in the Republic of Macedonia.

Keywords: drinking water, health, trihalomethanes, UV-VIS spectrophotometry.

INTRODUCTION

According to the World Health Organization (WHO), more than a billion people in the world have no access to potable water and more than three billion have a lack of adequate hygiene[1], therefore a very careful management of the drinking water needed. In this respect, the monitoring of chemical parameters in the determination of organic compounds in the drinking water is very important, since these compounds are harmful to human health[2]. Of these, highly dangerous are the disinfection byproducts (DBPs) whose main sub-group are trihalomethanes (THMs) which have proved to be cancerous to people. Having this in consideration, the level of awareness of the public opinion in relation to the quality of the drinking water, especially to the THMs has increased lately[3].

The reaction between chlorine and organic compounds present in the drinking water always produces THMs, when the former used as a disinfectant in advance. The presence of THMs in the final stage of the drinking water was discovered for the first time by Rook (1974)[4] in the City of Rotterdam, whereas Bellar et al (1974)[5] reported five months later that THMs had been found in the drinking water in the US.

Recently, the anxiety of healthcare authorities has been increasing due to the presence of the THMs in the drinking water, because of which harmful health effects emerge in consumers due to the constant and long-term exposal to the drinking water itself[6]. Waters that contain organic precursors (synthetic or natural) especially considered the main source of THMs. In many cases, the humic substances soluble in water are THM precursors; however, other organic substances such as algae and their extracellular products can be key precursors[7].

The scientific interest in THMs has increased since the cancerous features of THMs were recorded[8,9]. Because of this, certain countries have set maximal limits and guidelines when it comes to the THMs, which vary from one state to another (**Table-1**)[10]. Total THMs are determined as a sum of concentrations of chloroform, bromoform, bromodichloromethane and dibromochloromethane.

The aim of the article is to determine the variation of the THMs concentration and physical-chemical parameters in the drinking water in the city of Tetova during the summer season 2011, in order to conclude the quality of the drinking water and its impact on the health of the population living in this region.

EXPERIMENTAL

The city of Tetova is situated in the north-west part of Macedonia and has about 70 000 inhabitants. Even though it has sufficient water resources and permanent water flows, the lack of water felt in this city. The expenditure per household has decreased from 2.5 million m^3 in 1998 to 2 million m^3 in 2008[11]. The statistics show that the average amount of water per inhabitant is about 350-400 liters per day. The flow in the pipes of the reservoir is about 30 800 m^3/day. The drinking water in Tetova disinfected with gaseous chlorine without any kind of special treatment, whereas the South East European University (SEEU) utilizes the underground drinking water that extracted from three personal wells and it disinfected by a UV radiation used as primary disinfectant.

The experimental part of the research done in the laboratories of the State University of Tetova (SUT). Fifteen sample points selected in the city of Tetova (T1 – T15) and during the months of June, July and August 2011 the drinking water samples analyzed. The samples collected in glass bottles of 1.5 liters. Before collecting the water samples, a 1.5 mL solution of $Na_2S_2O_3$ 10% added in the bottles in order to remove the residual chlorine and to prevent the emergence of additional THMs.

The determined parameters were as follows: THMs, water temperature, turbidity, residual chlorine, pH, electrical conductivity (EC), the total residue after of evaporation (TRAE), total dissolved solids (TDS), chemical oxygen demand (COD), total organic carbon (TOC), dissolved organic carbon (DOC), ultra-violet absorbance in 254 nm (UV_{254}), specific ultra-violet absorbance (SUVA), nitrates and chlorides. Various different chemicals with pro-analysis, suprapur and HPLC cleanness were used.

The following instruments were used in the study: Portable Conductivity Meter WTW LF 320; portable pH-meter 330i WTW, turbidimeter, spectrophotometer UV-Vis Ultrospec and gas chromatograph Hewlett-Packard HP 5890 Series II ECD/FID and TOC Shimadzu analyzer.

The THMs in the drinking water are usually determined with the method of gas chromatography (GC). However, the majority of water management municipal services do not possess the adequate equipment, budget and professional cadres for this purpose. Therefore, the method of UV-VIS spectrophotometry was chosen for the quantitative determination of THMs, which requires instruments, freer reagents and shorter analysis period.

Table-1: Standards/Recommending guidelines for THMs (mg/L) in the world jurisdictions

Compound	WHO (1993)	USEPA (2001)	Health Canada (2001)	Aus – NZ (2000)	UK (2000)	EU (2001)
Chloroform	0.200	0.000*	–	–	–	–
Bromodichloromethane	0.060	0.060*	–	–	–	–
Dibromochloromethane	0.100	0.000*	–	–	–	–
Bromoform	0.100	0.000*	–	–	–	–
Total trihalomethanes	(THMs/WHO) ≤ 1**	0.080	0.100	0.250	0.100	0.100

****The maximum target level of pollution;* ***** *the sum of ratio of THMs levels guidance value should not exceed 1*

The determination of THMs using the method of UV-VIS spectrophotometry: Ten mL of pentane added in a normal dish containing 1L of drinking water to be analyzed. The dish shaken for about 3 minutes and then was left still until the two separate layers were visible. The pentane layer then removed and was added to a test tube containing 2 mL of NaOH 50% and 3 mL of pyridine. The test tube placed in a water bath at 45°C for 30 minutes in order to relieve the evaporation of pentane. Afterwards, the bath temperature was increased to 55°C for 45 minutes and later once more to 95°C for another 45 minutes[12,13]. After this, 2 mL of the pyridine layer (with a pink color) were removed and after the refrigeration was transferred to a 1 cm glass civet and the absorbance in 525 nm was measured (**Figure 1).**

One mL of bromoform and 1 mL of chloroform were added in 1 000 mL of methanol with the purpose of the construction of calibration curve. The total THMs concentration for this solution was 4.37 mg/mL and this was the initial standard solution of THMs. The standard solutions for the calibration curve with concentrations of 25, 50, 70, 100 and 125 μg/L were prepared by diluting the initial solution of THMs and each diluted by using 1 L of distilled water **(Figure 2)**.

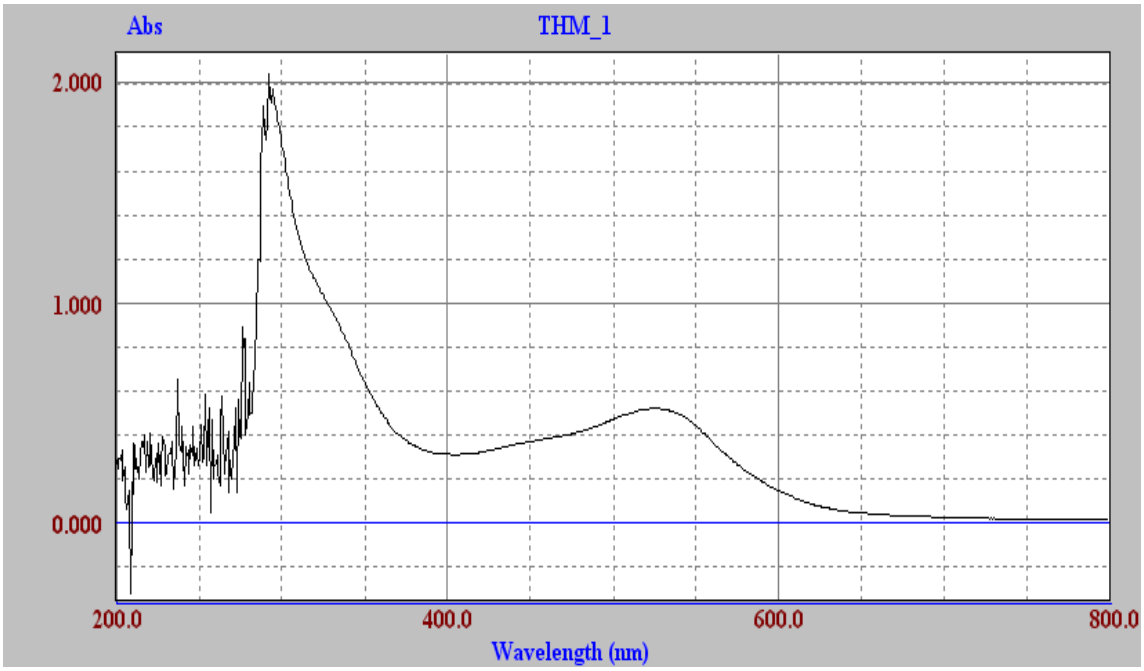

Figure 1: THMs UV/VIS absorption spectrum, γ= 50 µg/L

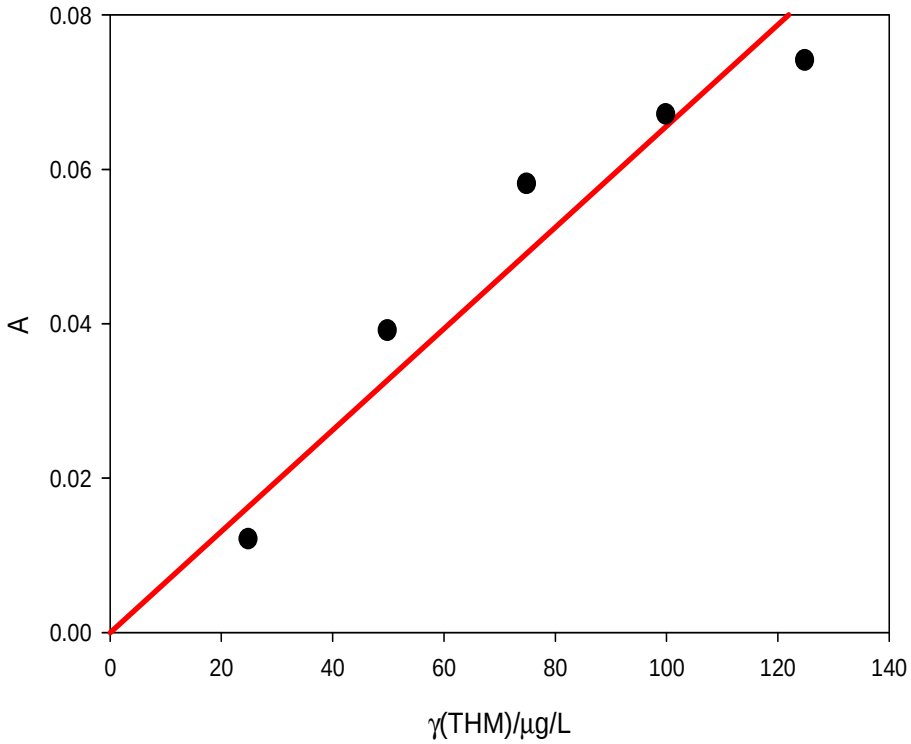

Figure 2: Calibration curve for THMs determination, July 2011

These solutions processed the same way as the samples of the drinking water were. This method based on the Fujiwar's reaction.

The determination of THMs with gas chromatography method carried out in accordance with the guidelines described in the literature[14].

RESULTS AND DISCUSSIONS

The experimental results have been presented in **Tables 2-4** and **Figures 1-4**. The concentration of THMs on the sample points vary from one month to another at a low rate (**Tables 2-4**). Therefore, the average values of THM concentrations during the months of June, July and August were 21.89, 25.70 and 26.66 µg/L respectively. Since the SEEU is supplied with underground water from wells and the water is disinfected only with UV radiation, during the period of measurements the residual chlorine was not detected, and as a consequence the concentration of THMs on the sample points T14 and T15 was 0.00 µg/L. If we neglect these two points, then the T1 sample point in June had the lowest value of 15.04 µg/L, whereas the T10 sample point had the highest value of 37.63 µg/L. In July, the lowest and highest values were recorded on T5 and T10 sample points with 17.81 µg/L and 42.73 µg/L respectively. In August, the sample point T5 had the lowest value of 16.63 µg/L and T10 had the greatest value of 45.08 µg/L (**Figure 3).**

Of the monthly averages on the sample point, it was concluded that the lowest average value of THMs in summer was detected on T5 with 17.02 µg/L, whereas the highest average value on T10 with 41.81 µg/L. During the whole summer season the range of THMs concentrations was 0.00 – 45.08 µg/L, whereas the average value with a standard deviation was 24.75 ± 12.33 µg/L. This concentration of THMs in Tetova's drinking water is under the recommended values of the National Regulations for the drinking water (100 µg/L)[15], which is harmonized with the recommendations by the WHO and the EU. From our preliminary research of THMs in Tetova's drinking water[16,17] we can conclude that the variation of THMs concentration in summer (24.75 ± 12.33 µg/L) was higher than that of the winter (17.41 ± 8.54 µg/L) and spring (20.06 ± 9.72 µg/L).

The concentration variation of THMs in the summer season was at its highest value in the month of August (26.66 µg/L) as a consequence of higher values of the following parameters: water temperature, pH, COD, TOC, DOC, residual chlorine, UV 254 and SUVA. The lower THMs values on T5, T1 and T6 refer to the shorter distance between the chlorination reservoir (short contact time), whereas the higher THMs values on T10, T11 and T9 refer to the longer distance between the chlorination reservoir (long contact time).

An additional factor for the emergence of higher values on the mentioned sample points can also be the organic pollution which can penetrate into the drinking water as a result of the outdated water supply system in the city, defects, frequent reparations as well as wastewaters. Other parameters of the quality of the drinking water will not be discussed in this article. They were measured for purposes of extracting mathematical models in order to predict the THMs concentrations in the drinking water in Tetova and are not a subject of this article.

The GC method is quite suitable and was also used for specifying the THMs in the drinking water in Tetova **(Figure 4).** For this purpose, in July, individual measurements of chloroform ($CHCl_3$), bromodichloromethane ($CHBrCl_2$), dibromochloromethane ($CHBr_2Cl$) and bromoform ($CHBr_3$) on T2, T6, T9, T10 and T11 were carried out. The results show that $CHBr_3$ were not detected on any of the sample points (0.00 µg/L), whereas $CHCl_3$ was the most represented species in the total THMs with a percentage from 78.65 % on T2 to 90.68 % on T10. $CHBrCl_2$ appeared with a very low percentage of 6.31 % on T10 and 12.31 % on T11. $CHBr_2Cl$ appeared with a very low percentage of 2.99 % on T10 and 8.35 % on T2. These results are compatible with those of other researchers and show that the most represented species in the total THMs is chloroform.

Table 2: Results of measurements and statistics, June 2011

Sample point	Tempe-rature	Turbi-dity	Residual chlorine	pH	EC	TRAE	TDS	COD	TOC	DOC	UV_{254}	SUVA	Nitrates	Chlorides	THMs
T1	10.60	1.30	0.1606	7.05	235.00	134.00	218.00	3.52	3.480	3.31	0.1120	0.03384	1.60	1.70	15.0360
T2	10.80	0.30	0.2300	7.15	265.00	137.00	172.00	2.68	2.560	2.48	0.0850	0.03427	1.10	1.80	23.6450
T3	10.90	0.30	0.2014	7.23	254.00	139.00	173.00	2.69	2.570	2.49	0.0860	0.03454	0.80	1.90	23.6450
T4	10.70	0.50	0.2107	8.16	278.00	145.00	177.00	2.72	2.580	2.51	0.0870	0.03466	0.80	2.50	28.1400
T5	10.80	0.30	0.3038	7.25	283.00	132.00	165.00	2.14	2.030	1.97	0.0670	0.03401	0.90	0.70	16.6320
T6	11.00	0.70	0.2600	7.31	285.00	139.00	173.00	2.95	2.840	2.73	0.0930	0.03407	0.40	1.80	19.2640
T7	10.80	0.80	0.1907	7.48	271.00	147.00	185.00	2.98	2.880	2.69	0.0910	0.03388	0.60	2.60	26.8310
T8	11.00	0.80	0.2425	8.06	273.00	145.00	187.00	3.06	2.970	2.81	0.0950	0.03381	1.10	2.70	26.8310
T9	10.70	1.10	0.1507	8.19	288.00	153.00	193.00	3.24	3.120	3.04	0.1030	0.03388	1.20	2.90	33.2640
T10	10.80	1.50	0.2544	8.34	293.00	157.00	226.00	3.87	3.620	3.52	0.1190	0.03381	1.40	3.60	37.6320
T11	11.10	1.50	0.2847	8.27	284.00	158.00	235.00	4.25	3.170	3.07	0.1040	0.03388	2.30	3.80	21.0000
T12	11.50	0.90	0.2201	8.26	266.00	142.00	192.00	2.85	2.680	2.53	0.0860	0.03399	1.30	2.40	25.6760
T13	11.20	1.30	0.2107	8.15	283.00	146.00	184.00	3.61	3.570	3.50	0.0840	0.02400	1.70	16.30	30.7720
T14	11.20	1.10	0.0000	7.14	664.00	362.00	491.00	3.82	3.660	3.52	0.0850	0.02415	21.50	24.30	0.0000
T15	11.20	1.10	0.0000	7.16	663.00	368.00	493.00	3.84	3.670	3.51	0.0860	0.02450	21.70	24.50	0.0000
Min	10.60	0.30	0.0000	7.05	235.00	132.00	165.00	2.14	2.03	1.970	0.067	0.02400	0.40	0.70	0.0000
Max	11.50	1.50	0.3038	8.34	664.00	368.00	493.00	4.25	3.67	3.520	0.119	0.03466	21.70	24.50	37.6320
Median	10.90	0.90	0.2107	7.48	283.00	145.00	187.00	3.06	2.97	2.810	0.087	0.03388	1.20	2.60	23.6450
Average	**10.95**	**0.90**	**0.1947**	**7.68**	**325.67**	**173.60**	**230.93**	**3.21**	**3.03**	**2.912**	**0.092**	**0.03209**	**3.89**	**6.23**	**21.8912**
Stan. Dev.	0.247	0.4192	0.0892	0.520	137.947	78.087	107.955	0.583	0.50	0.485	0.013	0.00408	7.2043	8.233	10.7075
Sum	164.3	13.50	2.9203	115.2	4885.00	2604.00	3464.00	48.22	45.40	43.68	1.383	0.48129	58.40	93.50	328.368
N	15	15	15	15	15	15	15	15	15	15	15	15	15	15	15

Sample points (with distance from the reservoir of chlorination) were: T1 – Faculty of Arts (4 km), T2 – Street 163 No. 11 (3.3 km), T3 – Butcher shop „Kadi çeshma“ (2.5 km), T4 – Primary school „Naim Frashëri“ (1.9 km), T5 – Teleferiku (0.8 km), T6 – Xhamia e larme (1.7 km), T7 – Eski xhamia (2.8 km), T8 – School of music (3.1 km), T9 – Xhamia Tabhane (3.4 km), T10 – NT “Atllantida” (4.1 km), T11 – Bus station (3.8 km), T12 - NT "Matica 2” (3.5 km), T13 – SUT (State University of Tetova – Faculty of Natural and Mathematical Sciences) (2.9 km), T14 – SEEU 1 (South East European University - Institute for Environment and Health) and T15 – SEEU 2 (South East European University - Canteen) (4.3 km).

Table 3: Results of measurements and statistics, July 2011

Sample point	Tempe-rature	Turbi-dity	Residual chlorine	pH	EC	TRAE	TDS	COD	TOC	DOC	UV_{254}	SUVA	Nitrates	Chlorides	THMs
T1	10.70	0.90	0.1070	7.26	241.00	153.00	184.00	4.36	4.270	4.14	0.1420	0.03430	0.80	1.50	19.9920
T2	10.90	0.20	0.1794	7.38	276.00	156.00	192.00	2.73	2.660	2.53	0.0500	0.01976	1.20	1.93	27.4140
T3	11.20	0.40	0.2107	7.39	278.00	159.00	194.00	2.74	2.670	2.54	0.0510	0.02008	1.20	1.94	27.4140
T4	10.90	0.40	0.2207	8.23	283.00	164.00	198.00	2.78	2.710	2.56	0.0520	0.02030	1.30	2.70	29.0360
T5	11.10	0.20	0.2501	7.08	237.00	152.00	182.00	2.34	2.260	2.14	0.0720	0.03640	1.00	0.80	17.8080
T6	11.30	0.80	0.2400	7.42	265.00	156.00	188.00	3.06	2.950	2.86	0.0970	0.03392	1.60	2.30	19.6840
T7	11.10	0.90	0.2600	8.13	281.00	167.00	193.00	3.14	3.020	2.87	0.0980	0.03415	2.20	2.10	30.9120
T8	11.20	0.60	0.2519	8.24	282.00	168.00	195.00	3.38	3.280	3.15	0.1070	0.03397	2.50	2.40	32.3680
T9	10.90	1.20	0.2226	8.56	291.00	172.00	197.00	3.41	3.350	3.27	0.1110	0.03394	2.40	2.90	37.9120
T10	11.10	1.40	0.1476	8.70	302.00	184.00	235.00	4.53	4.430	4.36	0.1480	0.03394	2.80	12.30	42.7280
T11	11.10	1.60	0.2300	8.82	294.00	173.00	246.00	4.67	4.540	4.41	0.1490	0.03379	2.70	12.80	40.9920
T12	11.40	1.00	0.2600	7.58	275.00	155.00	188.00	3.12	3.020	2.88	0.0960	0.03333	1.40	2.70	25.8160
T13	10.90	1.40	0.2712	8.34	280.00	168.00	202.00	3.45	3.270	3.15	0.1070	0.03397	2.30	18.60	33.4040
T14	11.10	0.90	0.0000	7.25	678.00	376.00	513.00	3.96	3.810	3.66	0.1240	0.03388	23.60	28.30	0.0000
T15	11.10	0.90	0.0000	7.28	675.00	378.00	515.00	3.98	3.820	3.67	0.1250	0.03406	23.80	28.10	0.0000
Min	10.70	0.20	0.0000	7.08	237.00	152.00	182.00	2.34	2.260	2.14	0.0500	0.01976	0.80	0.80	0.0000
Max	11.40	1.60	0.2712	8.82	678.00	378.00	515.00	4.67	4.540	4.41	0.1490	0.03640	23.80	28.30	42.7280
Median	11.10	0.90	0.2226	7.58	281.00	167.00	195.00	3.38	3.270	3.15	0.1070	0.03394	2.20	2.70	27.4140
Average	**11.07**	**0.85**	**0.1901**	**7.84**	**329.20**	**192.07**	**241.47**	**3.44**	**3.337**	**3.21**	**0.1019**	**0.03132**	**4.72**	**8.09**	**25.6987**
Stan. Dev.	0.180	0.434	0.0892	0.603	142.07	75.597	112.059	0.712	0.696	0.701	0.0336	0.00587	7.7332	9.7041	12.7418
Sum	166.0	12.80	2.8512	117.7	4938.00	2881.00	3622.00	51.65	50.060	48.19	1.5290	0.46979	70.80	121.37	385.480
N	15	15	15	15	15	15	15	15	15	15	15	15	15	15	15

Tabe 4: Results of measurements and statistics, August 2011

Sample point	Tempe-rature	Turbi-dity	Residual chlorine	pH	EC	TRAE	TDS	COD	TOC	DOC	UV_{254}	SUVA	Nitrates	Chlorides	THMs
T1	10.90	1.40	0.1182	7.36	254.00	162.00	189.00	4.59	4.410	4.28	0.1450	0.03388	1.20	1.80	17.9450
T2	11.20	0.30	0.1502	7.51	281.00	166.00	196.00	3.06	2.960	2.82	0.0950	0.03369	1.60	3.30	30.1840
T3	11.50	0.40	0.1502	7.52	282.00	167.00	197.00	3.08	2.970	2.83	0.0960	0.03392	1.80	3.50	31.6450
T4	11.20	0.40	0.1607	7.64	325.00	172.00	195.00	3.12	3.030	2.85	0.0970	0.03403	2.40	4.60	33.5440
T5	11.30	0.30	0.2501	7.12	258.00	158.00	184.00	2.67	2.540	2.46	0.0830	0.03374	1.30	1.10	16.6320
T6	11.60	0.70	0.2596	7.43	273.00	184.00	198.00	3.34	3.280	3.17	0.1070	0.03375	2.30	3.70	19.4040
T7	11.40	0.80	0.2807	8.05	291.00	185.00	201.00	3.42	3.380	3.26	0.1100	0.03374	3.80	3.90	30.1840
T8	11.50	0.90	0.3207	8.14	294.00	184.00	205.00	3.45	3.390	3.27	0.1200	0.03697	2.90	4.20	28.8870
T9	11.30	1.20	0.3113	8.53	302.00	197.00	208.00	3.52	4.440	4.38	0.1480	0.03379	3.50	8.30	39.0880
T10	11.40	1.50	0.2400	8.81	385.00	203.00	243.00	3.87	3.670	3.55	0.1200	0.03380	3.60	16.40	45.0800
T11	11.30	1.60	0.2501	8.76	420.00	231.00	265.00	3.64	3.540	3.47	0.1180	0.03401	3.70	18.30	43.5960
T12	11.60	1.10	0.2045	7.46	288.00	164.00	187.00	3.37	3.290	3.16	0.1070	0.03086	1.70	3.20	27.7200
T13	11.30	1.20	0.2200	8.52	296.00	177.00	227.00	3.82	3.660	3.54	0.1190	0.03362	2.70	19.40	36.0410
T14	11.40	1.30	0.0000	7.37	691.00	384.00	522.00	3.67	3.530	3.43	0.1160	0.03382	28.30	32.40	0.0000
T15	11.40	1.30	0.0000	7.38	693.00	386.00	526.00	3.68	3.540	3.42	0.1160	0.03392	28.50	32.60	0.0000
Min	10.90	0.30	0.0000	7.12	254.00	158.00	184.00	2.67	2.540	2.46	0.0830	0.03086	1.20	1.10	0.0000
Max	11.60	1.60	0.3207	8.81	693.00	386.00	526.00	4.59	4.440	4.38	0.1480	0.03697	28.50	32.60	45.0800
Median	11.40	1.10	0.2200	7.52	294.00	184.00	201.00	3.45	3.390	3.27	0.1160	0.03380	2.70	4.20	30.1840
Average	**11.35**	**0.96**	**0.1944**	**7.84**	**355.53**	**208.00**	**249.53**	**3.49**	**3.442**	**3.33**	**0.1131**	**0.03384**	**5.95**	**10.45**	**26.6633**
Stan. Dev.	0.177	0.4517	0.0988	0.574	143.65	74.292	113.594	0.444	0.504	0.514	0.0175	0.00116	9.1551	10.791	13.6863
Sum	170.3	14.40	2.9163	117.6	5333.00	3120.00	3743.00	52.30	51.630	49.89	1.6970	0.50754	89.30	156.70	399.950
N	15	15	15	15	15	15	15	15	15	15	15	15	15	15	15

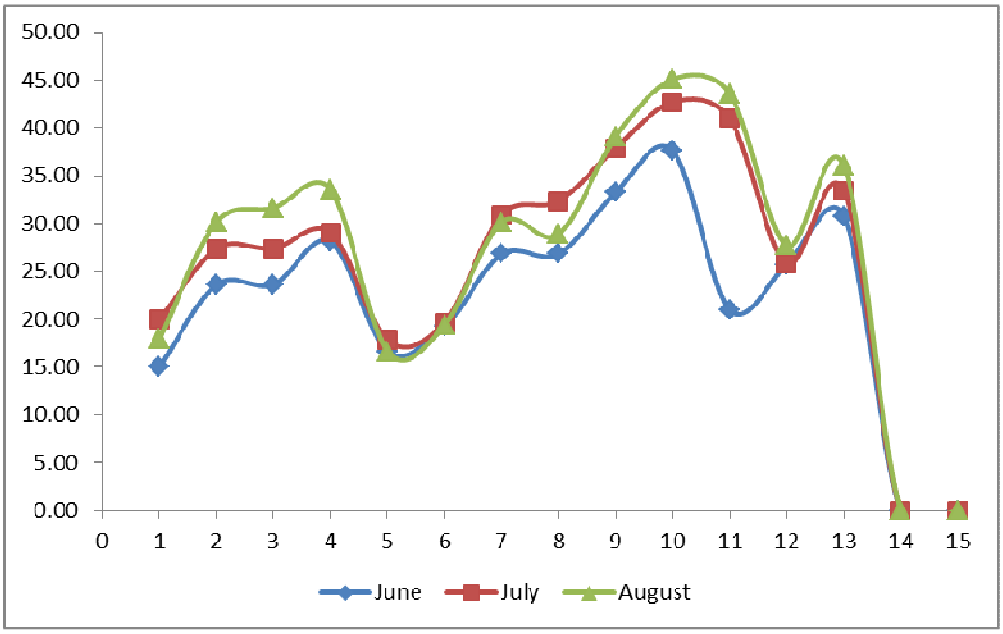

Figure 3: Spatial variation of THMs concentration in the summer season 2011

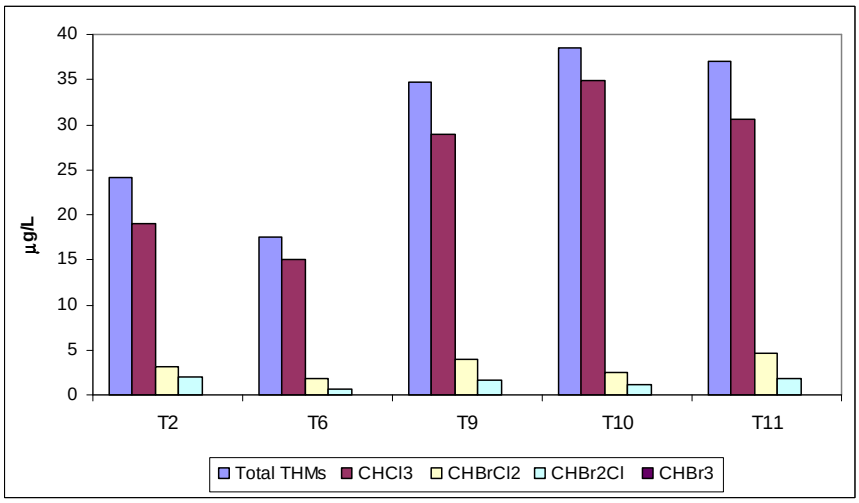

Figure 4: Gas chromatography analysis results of THMs, July 2011

CONCLUSIONS

The presence of THMs in the drinking water in the last decades has caused great worries since these compounds can cause cancer in humans. The monitoring of the THMs formation is crucial in order to make sure that the drinking water remains at the acceptable security levels. Therefore, the actions to reduce the THMs should be encouraged and there should be no compromise when it comes to the water disinfection.

The results of this study show that the level of THMs concentrations in the drinking water in the city of Tetova in the summer 2011 amounting at 24.75 ± 12.33 µg/L has been under the recommended values of the National Regulation for the drinking water, the WHO and the EU, and is currently safe for the population of this region. However, since the consumption of drinking water with THMs can cause health problems, we recommend to respective authorities to undertake preventive measures in keeping these values under control, especially when having in mind that during the hotter months the variation level of THMs can be very high.

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