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Seasonal Variation of Trihalomethanes Concentration in Tetova's Drinking Water (Part B)

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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 spring 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 was measured in fifteen different locations in March, April and May 2011. The results indicate that the seasonal variation is below the critical values stipulated in the state, EU, and WHO regulations (seasonal average 20.06 ± 9.72 µg/L.) This study is the first of its kind on THMs in the Republic of Macedonia. In conclusion, it can be inferred that the concentration of THMs in the drinking water of Tetova is not hazardous to human health.

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

1.0 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 (WHO, 2001), therefore a very careful management of the drinking wa-ter is needed. In this respect, the monitoring of che-mical parameters in the determination of organic compounds in the drinking water is very important, since these compounds are harmful to the human health (Durmishi et al, 2012). 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 aware-ness of the public opinion in relation to the quality of the drinking water, especially to the THMs has increased lately (Government of Newfounland and Labra-dor, 2000).

The reaction between chlorine and organic compounds present in the drinking water always produces THMs, when the former is 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) in the City of Rotterdam, whereas Bellar et al (1974) reported five months later that THMs had been found in the drinking water in the US.

Recently, the concern of healthcare authorities has been increasing due to the presence of the THMs in the drinking water, as a consequence of which harmful health effects emerge in consumers due to the constant and long-term exposal to the drinking water itself (Contu et al, 1990). Waters that contain organic precursors (synthetic or natural) are especially considered to be the main source of THMs. Generally, water soluble humic acids are THMs precursors (El-Shafy et al, 2000). After the discovery of carcinogenic properties, THMs are becoming very important (Symons et al, 1975; Young et al, 1987). Therefore, to protect the health some states have set the maximal limits and allowed guidelines for THMs, which vary from one state to another (Table 1) (Sadiq and Rodriguez, 2004). Total THMs are determined as a sum of concentrations of chloroform, bromoform, bromodichloromethane and dibromochloromethane.

The aim of the paper is to determine the variation of the THMs concentration and physical-chemical parameters in the drinking water in the city of Tetova during the spring 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.

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*

2.0 Materials and Methods:

The city of Tetova is situated in the north-western part of Macedonia and has permanent water flows, yet the lack of water is 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 (Jashari, 2010). The statistics show that the average amount of water per inhabi-tant 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 is disinfected with gaseous chlorine, without any other treatment, whereas the Southeast European University (SEEU) utilizes the underground drinking water that is extracted from three personal wells and it is disinfected by a UV radiation used as primary disinfectant.

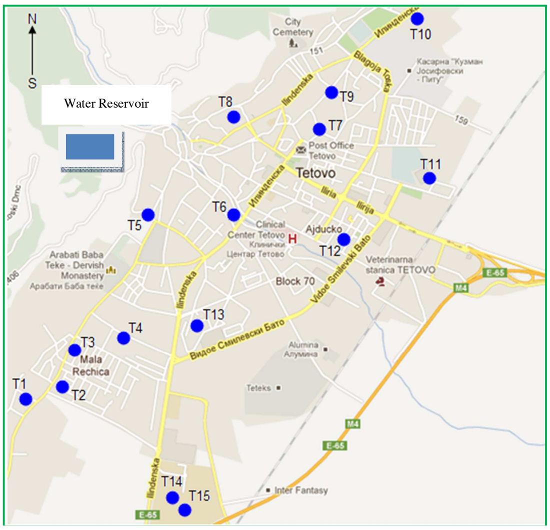

Fig 1: The map of the study area

The experimental part of this research was performed at the laboratories of the State University of Tetova (SUT). Fifteen sample were analyzed. The samples were collected in polyethylene bottles of 1.5 liters. Before collecting the water samples, a 1.5 mL soluti-on of $Na_2S_2O_3$ 10% was added in the bottles in order to remove the residual chlorine and to prevent the emergence of additional THMs.

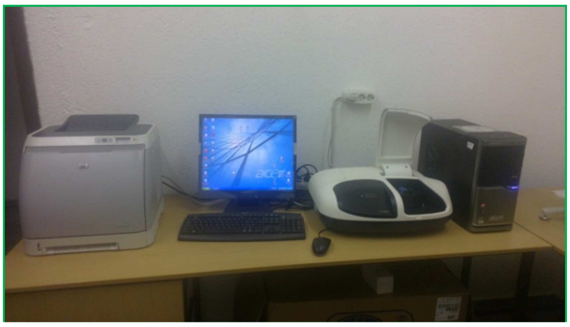

Fig 2: The photo of UV-VIS Spectrophotometer Ultrospec 5300 pro, Amersham Biosciences

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), che-mical 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 chlorines. 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, spectrophotometer UV-Vis HACH DR 2700, gas chromatograph Hewlett-Packard HP 5890 Series II ECD/FID and TOC Shimadzu analy-zer.

The THMs in the drinking water are usually determined with the method of gas chromatography (GC). However, the majority of water management municipal services does not possess the adequate equipment, budget and professional cadres for this purpose. Therefore, the method of UV-VIS spectropho-tometry was chosen for the quantitative determina-tion of THMs, which requires instruments, cheaper reagents and shorter analysis time frame. In order to see the result accuracy of the UV-VIS spectrophotometric method, certain results were compared to the UV-VIS spectrophotometric method developed by the HACH Company and is called the THM Plus Method and with GC, which are a very sensitive and precise methods. In this respect, during the month of May, the THMs were determined in five sample points using the three measurement methods.

2.1 The Determination of THMs Using Method of UV-VIS Spectrophotometry:

Ten mL of pentane were added in a normal dish containing 1L of drinking water to be analyzed. The dish was shaken for about 3 minutes and then was left still until the two separate layers were visible. The pentane layer was then removed and was added to a test tube containing 2 mL of NaOH 50% and 3 mL of pyridine. The test tube was 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 twice, at 55°C for 45 minutes and at 95°C for another 45 minutes (Huang et al, 1984; Espigares et al, 2003). 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 absor-bance in 525 nm was measured (Fig 3).

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 calibrating curve with concentrations of 25, 50, 70, 100 and 125 μg/L were prepared by diluting the initial solution of THMs and each was diluted by using 1 L of distilled water (Fig 4). These solutions were processed the same way as the samples of the drinking water. This method is based on the Fujiwar's reaction.

The determination of THMs with the UV-VIS spectro-photometric method according to HACH, was carried out in accordance with the procedures described by HACH Company, whereas the one using the GC method in accordance with guidelines described by Pauzi et al (2003).

3.0 Results and Discussion:

The experimental results have been presented in Tables 2-4 and Figures 3-7. 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 THMs concentrations during the months of March, April and May were 18.42, 20.84 and 20.92 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 dete-cted, 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 T5 sample point in March had the lowest value of 12.26 μg/L, whereas the T10 sample point had the highest value of 27.89 μg/L. In April, the lowest and highest values were respectively recorded on T1 and T10 sample points with 10.78 μg/L and 33.40 μg/L respe-ctively. In May, the sample point T1 had the lowest value of 6.15 μg/L and T10 had the greatest value of 32.25 μg/L.

Of the monthly averages on the sample point, it was concluded that the lowest average value of THMs in spring was detected on T1 with 11.33 μg/L, whereas the highest value on T10 with 31.18 μg/L. During the whole spring season the range of THMs concentrations was 0.00 – 33.40 μg/L, whereas the average value with a standard deviation was 20.06 ± 9.72 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), which is harmonized with the recommendations by the WHO and the EU.

The concentration variation of THMs in the spring season was at its highest value in the month of May (20.92 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 T1, T5 and T6 refer to the shorter distance between the chlorination reservoir (short contact time), whereas the higher THMs values on T10, T9, T11 and T13 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 in the system itself, frequent repairs as well as wastewaters mistreatment.

Other parameters of the quality of the drinking water will not be discussed in this paper. 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 research.

For verification the validity of the results, in May the THMs measurements were carried out with the three methods on sample points T3, T5, T8, T10 and T12 (Fig **6**). From the results it was resulted that the determination of THMs with UV-VIS spectrophotometrc method by HACH and GC method (methods 2 and 3) was more sensitive in comparison with UV-VIS spectrophotometrc method (method 1). Lower concentration of THMs was in sample point T5, and higher in T10. Thus, concentrations of THMs in sample point T5 with method 1, 2 and 3 were 19.97, 18.25 and 18.06 µg/L respectively, while those in sample point T10 were 32.25, 29.15 and 28.75 µg/L respectively. So, the order of methods sensitivity was: method 3 > method 2 > 1 method. Results of method 1 resulted in slightly higher than those of method 3 and 4, but they are very similar to those of method 2 and 3. . In order to increase the sensitivity and accuracy of the results by using method no. 1, we used reagents and solvents of HPLC cleanness. We can conclude that the UV-VIS spectrophotometric method can successfully be used in determining the THMs and it can replace methods no. 2 and 3.

Table 2: Results of measurements and statistics, March 2011

Sample point	Tempe-rature	Turbi-dity	Residual chlorine	pH	PE	TRAE	TDS	COD	TOC	DOC	UV_{254}	SUVA	Nitrates	Chlorines	THMs
T1	9.50	1.20	0.1032	7.48	202.00	124.00	108.00	2.52	2.380	2.26	0.0580	0.02566	0.700	1.200	17.0480
T2	9.40	0.80	0.1531	7.41	200.00	117.00	112.00	2.24	2.170	2.07	0.0320	0.01546	0.600	0.900	19.6820
T3	9.70	0.60	0.2000	7.46	199.00	115.00	110.00	2.21	2.100	1.96	0.0290	0.01479	0.500	0.800	19.6820
T4	9.80	0.80	0.2094	7.53	223.00	113.00	118.00	2.38	2.240	2.14	0.0420	0.01963	0.500	1.000	26.2640
T5	9.30	0.30	0.1500	7.46	221.00	102.00	116.00	2.35	2.210	2.08	0.0320	0.01538	0.300	0.600	12.2610
T6	9.50	0.50	0.1901	7.66	220.00	115.00	114.00	2.32	2.190	2.06	0.0310	0.01548	0.400	0.900	15.1760
T7	9.60	0.60	0.1652	7.22	222.00	121.00	121.00	2.54	2.380	2.27	0.0600	0.02643	0.600	1.200	21.7280
T8	9.70	1.10	0.2182	6.94	221.00	125.00	119.00	2.56	2.470	2.36	0.0690	0.02924	0.800	1.400	18.2280
T9	9.70	1.20	0.1805	7.04	223.00	130.00	115.00	2.63	2.520	2.43	0.0820	0.03374	1.200	1.500	24.4700
T10	9.60	1.40	0.2525	6.95	222.00	127.00	120.00	2.72	2.570	2.45	0.0810	0.03306	1.100	1.600	27.8910
T11	9.80	1.00	0.2418	7.26	222.00	132.00	121.00	3.52	3.260	3.16	0.1070	0.03386	1.300	1.600	24.4700
T12	9.70	1.20	0.2107	6.96	223.00	134.00	114.00	2.54	2.370	2.18	0.0440	0.02018	1.200	1.800	23.1850
T13	9.50	0.80	0.1805	7.15	222.50	110.00	118.00	2.33	2.140	2.03	0.0300	0.01478	2.300	12.000	26.2640
T14	9.90	0.90	0.0000	6.85	678.00	365.00	354.00	3.82	3.680	3.56	0.1260	0.03539	7.600	15.300	0.0000
T15	9.90	0.90	0.0000	7.11	680.00	368.00	357.00	3.83	3.690	3.57	0.1270	0.03557	7.400	14.900	0.0000

Min	9.30	0.30	0.0000	6.85	199.00	102.00	108.00	2.21	2.100	1.96	0.0290	0.01478	0.300	0.600	0.00
Max	9.90	1.40	0.2525	7.66	680.00	368.00	357.00	3.83	3.690	3.57	0.1270	0.03557	7.600	15.300	27.8910
Median	9.70	0.90	0.1805	7.22	222.00	124.00	118.00	2.54	2.380	2.26	0.0580	0.02566	0.800	1.400	19.6820
Average	**9.64**	**0.89**	**0.1637**	**7.23**	**278.57**	**153.20**	**147.80**	**2.70**	**2.558**	**2.44**	**0.0633**	**0.02458**	**1.767**	**3.780**	**18.4233**
Stan. Dev.	0.176	0.3044	0.0762	0.255	162.816	87.038	84.4158	0.55	0.537	0.54	0.035	0.00842	2.3811	5.3775	8.66752

Table 3: Results of measurements and statistics, April 2011

Sample point	Tempe-rature	Turbi-dity	Residual chlorine	pH	PE	TRAE	TDS	COD	TOC	DOC	UV_{254}	SUVA	Nitrates	Chlorines	THMs
T1	10.80	1.40	0.1495	7.51	233.50	129.00	185.00	2.61	2.530	2.44	0.081	0.03319	1.30	0.80	10.7800
T2	10.60	0.40	0.3506	7.73	223.50	122.00	160.00	2.32	2.110	2.05	0.031	0.01512	0.80	2.20	20.2760
T3	10.10	0.40	0.3406	7.48	200.00	120.00	160.00	2.34	2.260	2.11	0.039	0.01848	0.70	2.30	25.2300
T4	10.20	1.20	0.2019	7.52	201.00	118.00	160.00	2.36	2.280	2.13	0.041	0.01925	0.70	2.50	28.5860
T5	10.30	0.30	0.3475	7.25	198.50	112.00	155.00	2.04	1.950	1.87	0.063	0.03369	0.90	2.10	16.0440
T6	10.10	0.80	0.3031	7.31	200.50	119.00	160.00	2.37	2.220	2.14	0.042	0.01963	1.10	2.40	17.4800
T7	10.30	1.10	0.3481	7.04	199.00	131.00	160.00	2.38	2.240	2.15	0.043	0.02000	1.30	2.40	26.6820
T8	10.30	1.20	0.3537	6.62	201.00	128.00	160.00	2.45	2.340	2.26	0.058	0.02566	1.40	2.70	25.2300
T9	10.40	1.40	0.3570	6.79	199.50	126.00	160.00	2.48	2.370	2.31	0.062	0.02684	1.70	2.90	30.1830
T10	10.40	1.40	0.3093	6.93	200.50	132.00	160.00	2.53	2.390	2.32	0.063	0.02716	1.70	3.10	33.4000
T11	10.50	1.60	0.2957	6.62	200.00	135.00	160.00	2.55	2.430	2.37	0.069	0.02911	2.50	3.30	28.5860
T12	10.50	1.80	0.3107	6.89	199.00	137.00	155.00	2.62	2.550	2.48	0.084	0.03387	2.30	3.60	23.4930
T13	10.30	1.90	0.3012	7.08	222.00	139.00	155.00	2.81	2.660	2.53	0.086	0.03399	3.40	5.20	26.6820
T14	10.50	1.10	0.0000	7.09	685.00	372.00	485.00	2.31	2.240	2.13	0.041	0.01925	16.50	22.40	0.0000
T15	10.40	1.10	0.0000	7.03	684.00	371.00	485.00	2.33	2.260	2.15	0.042	0.01953	16.30	22.50	0.0000

Min	10.10	0.30	0.0000	6.62	198.50	112.00	155.00	2.04	1.950	1.87	0.031	0.01512	0.70	0.80	0.0000
Max	10.80	1.90	0.3570	7.73	685.00	372.00	485.00	2.81	2.660	2.53	0.086	0.03399	16.50	22.50	33.4000
Median	10.40	1.20	0.3093	7.08	200.50	129.00	160.00	2.38	2.280	2.15	0.058	0.02566	1.40	2.70	25.2300
Average	**10.38**	**1.140**	**0.2646**	**7.126**	**269.80**	**159.40**	**204.00**	**2.433**	**2.322**	**2.229**	**0.056**	**0.02498**	**3.507**	**5.36**	**20.8435**
Stan. Dev.	0.186	0.490	0.1221	0.336	168.728	86.443	114.302	0.179	0.178	0.179	0.018	0.007	5.287	7.001	10.293

Tabe 4: Results of measurements and statistics, May 2011

Sample point	Tempe-rature	Turbi-dity	Residual chlorine	pH	PE	TRAE	TDS	COD	TOC	DOC	UV_{254}	SUVA	Nitrates	Chlorines	THMs
T1	10.40	1.60	0.1507	6.94	281.00	121.00	225.00	2.58	2.490	2.33	0.0640	0.02747	1.20	2.40	6.1500
T2	10.50	0.20	0.2495	7.07	199.50	118.00	160.00	2.41	2.270	2.17	0.0730	0.03364	0.80	0.90	21.6160
T3	10.70	0.20	0.2014	7.06	200.50	117.00	160.00	2.37	2.280	2.18	0.0740	0.03394	0.60	1.10	25.6760
T4	10.60	0.40	0.1550	6.99	200.50	117.00	160.00	2.42	2.290	2.19	0.0750	0.03425	0.60	1.50	27.7200
T5	10.50	0.30	0.3013	7.34	199.00	115.00	160.00	2.16	1.880	1.78	0.0600	0.03371	0.20	0.60	17.8080
T6	10.80	0.80	0.2519	7.51	199.00	122.00	155.00	2.31	2.160	2.07	0.0320	0.01546	0.40	0.40	19.9960
T7	10.60	0.60	0.2408	7.07	200.00	124.00	160.00	2.45	2.170	2.09	0.0330	0.01579	0.70	2.20	25.6760
T8	10.80	0.60	0.2501	6.08	200.50	128.00	160.00	2.46	2.380	2.24	0.0560	0.02500	0.80	2.40	24.8080
T9	10.50	1.20	0.2607	6.33	202.00	126.00	160.00	2.52	2.390	2.25	0.0580	0.02578	0.90	2.80	30.6320
T10	10.60	1.20	0.2107	6.54	201.50	128.00	160.00	2.62	2.420	2.33	0.0640	0.02747	1.30	2.99	32.2530
T11	10.80	1.40	0.3000	7.03	199.50	132.00	160.00	2.59	2.430	2.34	0.0650	0.02778	1.20	3.20	28.8940
T12	11.30	1.20	0.2346	8.34	197.50	134.00	155.00	2.73	2.570	2.46	0.0830	0.03374	2.10	3.20	24.8080
T13	10.70	1.50	0.2063	8.24	203.00	136.00	160.00	2.93	2.640	2.57	0.0870	0.03385	2.50	13.40	27.7200
T14	11.20	1.20	0.0000	7.02	652.00	358.00	495.00	3.84	3.610	3.51	0.1180	0.03362	18.20	21.30	0.0000
T15	11.10	1.20	0.0000	7.08	651.00	357.00	490.00	3.85	3.590	3.50	0.1170	0.03343	18.10	21.20	0.0000
Min	10.40	0.20	0.0000	6.08	197.50	115.00	155.00	2.16	1.880	1.78	0.0320	0.01546	0.20	0.40	0.0000
Max	11.30	1.60	0.3013	8.34	652.00	358.00	495.00	3.85	3.610	3.51	0.1180	0.03425	18.20	21.30	32.2530
Median	10.70	1.20	0.2346	7.06	200.50	126.00	160.00	2.52	2.390	2.25	0.0650	0.03343	0.90	2.40	24.8080
Average	**10.74**	**0.907**	**0.2009**	**7.11**	**265.77**	**155.53**	**208.00**	**2.683**	**2.505**	**2.40**	**0.0706**	**0.02900**	**3.31**	**5.31**	**20.9171**
Stan. Dev.	0.269	0.4877	0.0922	0.601	157.981	82.246	116.754	0.51	0.481	0.483	0.024	0.0064	6.0567	7.1654	10.5518

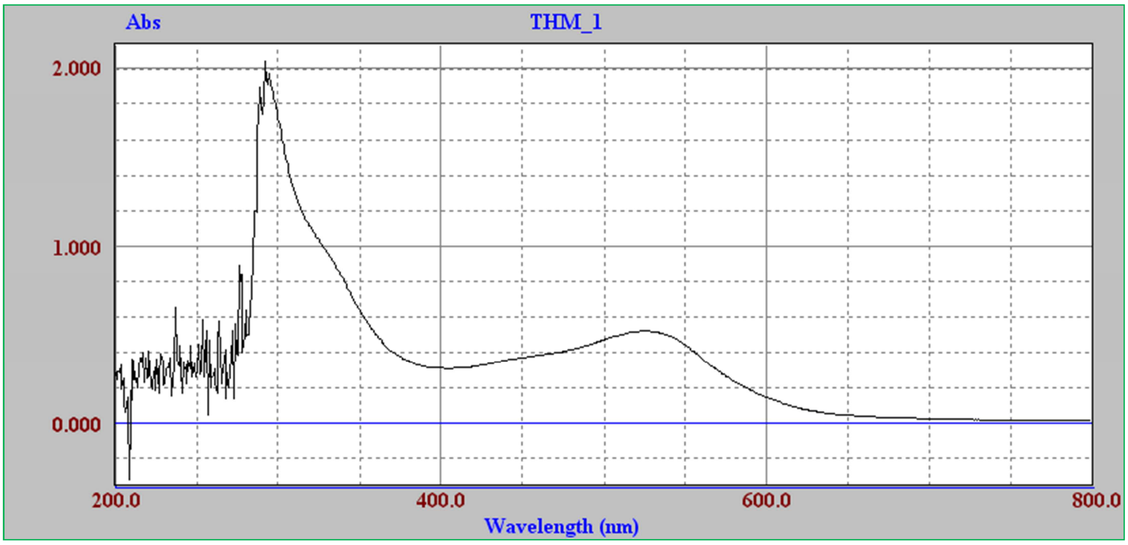

Fig 3: THMs UV/VIS absorption spectrum, γ = 50 μg/L

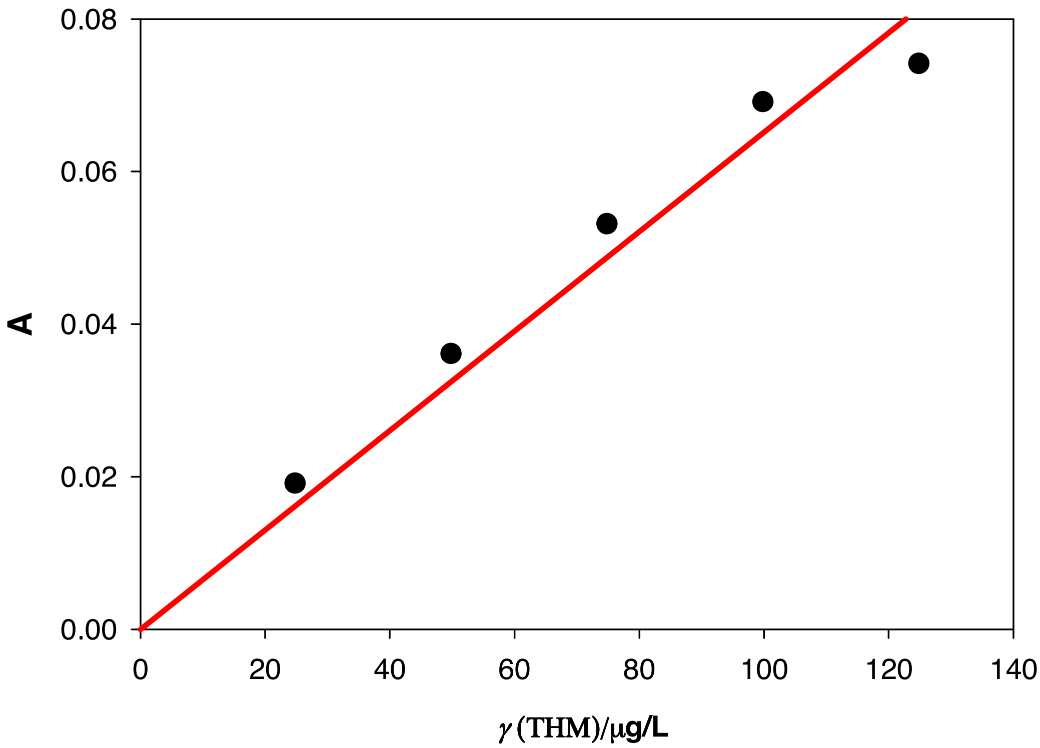

Fig 4: Calibration curve for THMs determination, May 2011

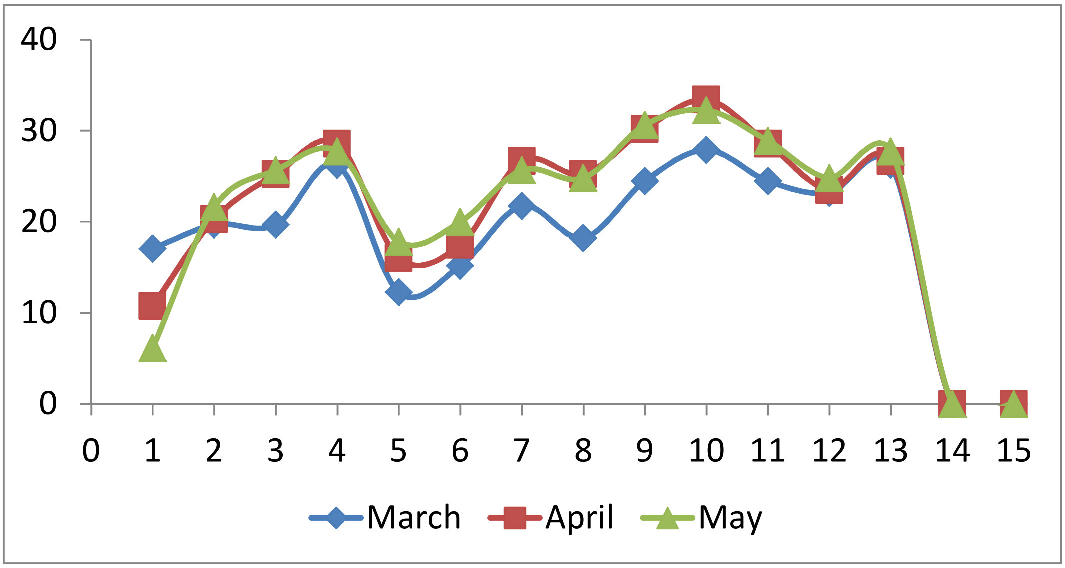

Fig 5: Spatial variation of THMs concentration in the spring season 2011

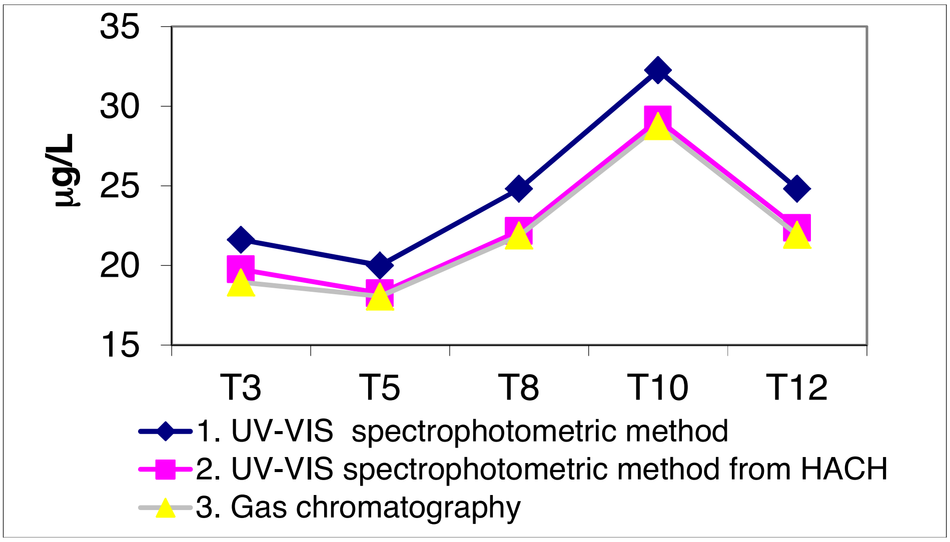

Fig 6: Comparison of the THMs results with three determination methods, May 2011

The GC method is quite suitable and was also used for specifying the THMs in the drinking water in Tetova (Fig 7). For this purpose, in May, individual measurements of chloroform ($CHCl_3$), bromodichlo-romethane ($CHBrCl_2$), dibromochloromethane ($CHBr_2Cl$) and bromoform ($CHBr_3$) on T3, T5, T8, T10 and T12 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 repre-sented specie in the total THMs with a percentage from 84.86 % on T12 to 92.37 % on T8. $CHBrCl_2$ appeared with a very low percentage of 4.43 % on T8 and 11.74 % on T5. $CHBr_2Cl$ appeared with a very low percentage of 0.00 % on T5 and 4.96 % on T3. These results are compatible with those of other researchers and show that the most represented specie in the total THMs is chloroform.

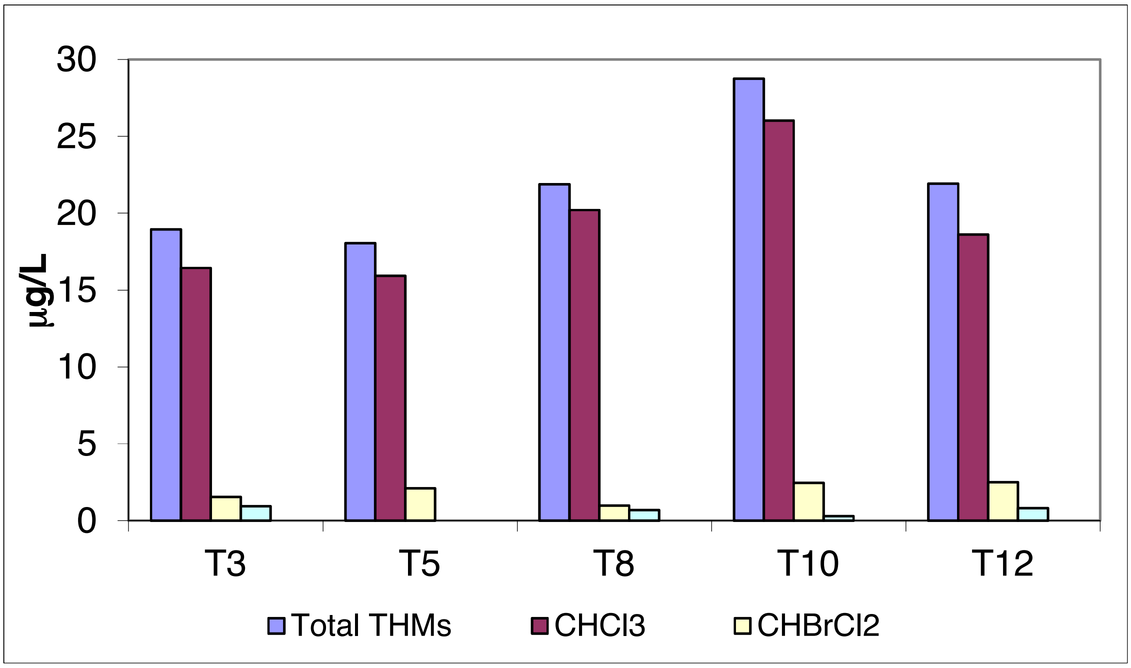

Fig 7: Gas chromatography analysis results of THMs, May 2011

4.0 Conclusions:

The presence of THMs in the drinking water in the last decades has caused great worries since these components 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 safety 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 variation of the THMs level shows the effect of different factors on their formation under the conditions of water chlorination as well as the performance of enterprises that deal with water treatment and management.

The results of this study show that the level of THMs concentrations in the drinking water in the city of Tetova in the spring 2011 amounting at 20.06 ± 9.72 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. Based on the research results, we can also conclude that the UV-VIS spectro-photometric method can successfully be applied in determining the THMs and it can replace the UV-VIS spectrophotometric method based on HACH as well as the gas chromatographic method.

5.0 Acnowledgment:

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