APPLICATION OF MATHEMATICAL MODELS FOR PREDICTING THE TRIHALOMETHANES CONTENT IN DRINKING WATER IN THE CITY OF DEBAR, NORTH MACEDONIA

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Abstract

Trihalomethanes (THMs) as the main disinfection byproducts created when chlorine reacts with the organic matter of the drinking water. THMs in high concentrations are harmful and can be carcinogenic for the liver, pancreas, nervous system, and development organs, whereas in women can cause miscarriage. Consequently, THMs must be constantly monitored. THMs mainly are determined by the gas chromatography method, which is a difficult procedure and very costly. To avoid this, in the past years, the use of mathematical models for the prediction of THMs in drinking water has been practiced. By fast measuring the values of some simple parameters of drinking water quality and replacing them in the mathematical models we can predict the THMs content. The aim of this article was the predict the THMs content in drinking water in the city of Debar for the spring of 2021 in four sampling points D1, D2, D3, and D4. The measured parameters were: water temperature, residual chlorine, pH, electrical conductivity, chemical oxygen demand, total dissolved solids, and chlorides. For prediction were used ten mathematical models and the average value of THMs with standard deviation was 23.88 ± 8.16 µg/L. From the results, we can conclude that the used models for THMs prediction have been successful and this content of THMs poses no risk to public health.

Keywords: THMs, physico-chemical parameters, drinking water, mathematical models for prediction, health

1. Introduction

THMs as synthetic organic compounds are created by the replacement of three hydrogen atoms in the methane molecule with the atoms of the halogen elements. Chloroform or trichloromethane (CHCl3), bromochloromethane (CHBrCl2), dibromochlormethane (CHBr2Cl), and bromoform or tribromomethane (CHBr3) can be formed during chlorination. Their formation during chlorination represents a very serious health problem as chloroform, the main subspecies of THMs formed in this process, is implicated in several types of cancers in laboratory animals (Durmishi, 2013).

In recent years, modeling for predicting of THM concentration has been the contemporary trend. Relevant models are developed with adequate statistical processing of THMs data with the help of statistical programs. Statistical software used for this purpose is Statistical Package for the Social Sciences (SPSS), Statgraphics, etc. The models confirm the empirical or mechanical correlation between THMs contents in drinking water and water quality parameters and their control may be related to the formation of THMs. These models are applied based on routine measurements of some drinking water parameters and with their help predict (calculate) the concentration of THMs in drinking water. These models are user-friendly and can be used by any worker in the drinking water plant to estimate the concentration of THM at any given time.

Several models have been developed to predict mathematically the total THMs (TTHMs) from the water source characteristics. This enables the calculation of TTHMs without the need for intensive sampling. *Golfinopoulos et al.* developed a model for TTHMs depending on chlorophyll a, pH, bromine concentration, season, temperature, and chlorine concentration (Golfinopoulos *et al.*, 1998). The model

showed that the concentration of TTHMs increases with the concentration of chlophyl a, bromine ion concentration, temperature, chlorine concentration, and whether the samples were taken in spring or summer. According to him, TTHM concentration decreases with the pH and temperature of the samples in the spring and summer seasons. Rodriquez and Serodes developed a model for predicting TTHMs in finished water in three plants and TTHMs produced at distribution system points (Rodriguez and Serodes, 2001). In two plants it was found that the temperature was the only significant variable and TTHM concentration increased with temperature. For the third plant the temperature, pH, and flow rate were important variables. It was noted that the concentration of TTHMs increased with temperature, and decreased with pH and flow. *Villanova et al.*, developed a model in two Torres River water treatment plants in the city of Salamanca, Spain (Villanueva *et al.*, 2006). This one-year research showed that temperature and pH were the only variables important for the formation of chloroform. Observed results versus calculated values had a high value of R2 = 0.99. As we can see, simple and some very complicated models for the THMs prediction have been developed.

Statistical analysis of results with multifactor variance analysis has revealed the influence of the parameters in the formation of THMs. Simple and multiple regression were used to develop predictive models for THMs formation. These models serve to predict concentrations of THMs in drinking water after monitoring the THMs and following relevant experiments on the impact of various factors on THMs concentration in drinking water. Each developed model is suitable only for drinking water with similar characteristics. So, a similar model cannot be used to predict THMs in drinking water of different locations with different characteristics and qualities (Durmishi, 2013).

Thus, Elshorbagy has modeled the formation of different THMs under strictly extreme conditions of chlorine concentration, temperature, and bromine ion concentration (Elshorbagy, 2000). Clark and Mano developed a mathematical model that predicts THMs concentration as a function of pH, temperature, initial chlorine concentration, and total organic carbon (TOC) (Clark and Sivaganesan, 1998). Montgomery Watson Consulting Engineering modeled the THMs formation associated with TOC, pH, temperature, chlorine concentration, bromine ion concentration, and contact time (Montgomery, 1993). Some other researchers such as *Clark et al.*, studied the effect of the bromine ion concentration on THMs formation (Clark *et al.*, 1996). Other researchers studying the effect of other factors on THMs formation. Karimi and Singer reported a strong correlation between algae productivity and THM formation potential (THMFP) (Karimi and Singer, 1991). *Canale et al.*, link THMFP with chlorophyll, zooplankton, water depth, dissolved oxygen, and total phosphorus (Canale *et al.*, 1997). The aim of this paper was the prediction of the THMs content in drinking water in the city of Debar for the spring 2021.

2. Materials and Methods

2.1 Sampling stations, periods, and sampling: Drinking water sampling stations are appropriately selected. Four stations (D1 - D4) have been designated in the city of Debar. A serious account has been taken of the distance between the stations to cover a certain part of the city's territory as well as possible and to enable logical conclusions to be drawn. The stations were: D1 - General Hospital; D2 -Mosque of Namazjar, D3 - Neighborhood "September 8" and D4 - Neighborhood "Qernanica". From the mentioned stations, drinking water samples were taken every week in March, April, May, and June of 2021 (spring season) to determine the values of some of the most necessary drinking water quality parameters. These parameters have been imperative for predicting the concentration of THMs with mathematical models.

The method of sampling has a great influence on the obtained results of the analyses. Sampling was done according to the recommendations of the State Regulation on drinking water of the Republic of North Macedonia, which is harmonized with WHO and EU recommendations (Government of the Republic of Macedonia, 2004). Drinking water samples were taken in polyethylene and glass bottles with a volume of 1.5 L. We filled the bottles up to the lid without leaving space and air bubbles in the sample.

2.2 *Measured parameters, reagents, and instruments:* In this paper, in the spring season, the monitoring of the seven most important drinking water parameters was carried out: water temperature (WT), residual chlorine (RC), pH, electrolytic conductivity (EC), total dissolved solids (TDS), chemical oxygen demand (COD) and chlorides.

The experimental part of the paper was carried out in the field, while COD and chlorides were measured in the laboratories of the University of Tetova. For the realization of this research, relevant methodology has been used and standard methods and techniques of measuring parameters have been used by the State Regulation for drinking water. Standard physicochemical methods were used to determine the parameters. The following reagents were used to measure the parameters: 1) tetra-methyl-benzidine (reagent for residual chlorine), 2) buffer solutions 4, 7 and 9 (for pH-meter calibration), 3) standard KCl solution (for calibration of the conductometer), 4) H₂SO₄ solution (1:3), 5) KMnO₄ solution with c = 0.002 mol/dm³, 6) H₂C₂O₄ solution with c = 0.002 mol/dm³, 7) 10% K₂CrO₄ solution (indicator for determination of chlorides) and 8) AgNO₃ solution with c = 0.0281 mol/dm³.

The following equipment and instruments were used to determine the physico-chemical parameters: a thermometer was used to measure the temperature (an integral part of the conductometer), a portable Conductivity Meter, WTW LF 320, was used to measure EC and TDS; pH measurement was done with a portable pH meter 330i, WTW; RC was measured colorimetrically with a comparator; chlorides were determined by argentometric titration and COD was determined by standard procedure using oxidizing reagents KMnO₄ and $H_2C_2O_4$.

2.3 Drinking water of the city of Debar: According to the last census, the city of Debar had 21 500 inhabitants. Previously, the city was supplied with water from Mount Deshat with a capacity of 60-80 L/s. Later, the city was supplied with water from Lake Debar through pumps of 60 L/s. Over time, the water of the lake became polluted and as a solution for water supply, the source of the village of Rosok (20 km from the city) with very large capacities was proposed. The minimum of this source is 480 L/s, while the maximum is 5500 L/s. From this source, the city of Debar receives a sufficient quantity of 200 L/s of water for the inhabitants of the city and the surrounding villages.

The Rosok aqueduct was launched in 1988. The length of this aqueduct is 19.557 km from Rosok to the reservoir of the upper area of the city, while the length of the pipeline is 70 km of plastic material and 355 mm of metal, which has internal and external isolation. The water pipeline has auxiliary facilities, 34 release valves and 34 air valves. This aqueduct was built with a value of \$2 million from its contribution of 2.5% of the citizens in 4 years. The water supply of Rosok, based on physical-chemical-bacteriological analyses, meets the norms for water utilization in all aspects.

This water supply works with free fall from the altitude of 1012 m above sea level to the reservoir of the upper area of 788 m. It is called a caustic spring and it comes out under pressure from the depth of the earth, which in certain cases when there is rain with great intensity and with a weekly duration, it can happen to wash the underground rocks. In these cases, the limestone dust causes the water to become cloudy, which is not physically harmful to the residents. Rosok's aqueduct has an optimal composition of dust, Mn, Fe, Si, etc. in an optimal percentage, which is considered one of the most qualitative sources in Europe.

The city of Debar has a reservoir in the upper area with a capacity of 500 m³, a reservoir in a quota of 712 m with a capacity of 750 m³ and a reservoir in a quota of 720 m with a capacity of 150 m³. Rosok water does not need chlorination, it can be consumed as it is, but for safety, chlorination is done every day with a quantity of 0.1; 0.2; up to 0.3 mg/L because different bacteria can appear from the city network.

3. Results and Discussion

The results of the measurements of this paper are presented in Figures 1 - 7 and Table 1.

3.1 Water Temperature (WT): Temperature plays a crucial role in the physicochemical and biological behavior of the water system (Dwivedi and Santoshi, 2004). Chemical reactions depend on water temperature and it controls the metabolic and reproductive processes of aquatic species. The water samples analyzed from the drinking water of the city of Debar had approximate temperatures and were part of the recommended value of the State Regulation. The range for WT was 9.00 - 12.00 °C. The lowest temperature of 9.00 °C was found in March in D3, while the highest temperature of 12.0 °C was found in June in D2, D3 and D4. The average water temperature values in March, April, May, and June were 10.25, 10.5, 10, and 11.75 °C respectively. The average seasonal value with standard deviation was 10.625 ± 0.78 °C, which was by the state regulation (Fig. 1).



Figure 1. Spatial and temporal variation of the WT

3.2 Residual Chlorine (RC): RC is of great importance to determine the presence or absence of microorganisms in drinking water. Its presence in drinking water indicates that a sufficient amount of chlorine has been added to the water first to inactivate bacteria and some viruses that cause diseases such as diarrhea and also to protect the water from recontamination during storage. The range for RC was 0.20 - 0.20 mg/L (Fig. 2). The average values in March, April, May and June were 0.20 mg/L, respectively. The average seasonal value with standard deviation was $0.20\pm0.00 \text{mg/L}$, which was by the state regulation.



Figure 2. Spatial and temporal variation of the RC

3.3 pH Value: The pH value of aquatic ecosystems depends on the chemical and biological activity of the water. Natural waters usually have a pH value higher than 7. This results from CO2 from the atmosphere and from that which is released from the decomposition of organic matter as well as from human activity. CO2 dissolves in water and forms H2CO3. This acid acts with CaCO3 of surface water and forms CaHCO3 and as a result, natural waters have a value of pH>7. The water samples that we analyzed had pH values in a range of 7.68 - 7.92. The lowest value was in June in D2, while the highest value was in May in D4. The average values in March, April, May, and June were 7.87; 7.7675; 7.8875, and 7.765 respectively (Fig. 3). The average seasonal value with standard deviation was 7.8225 \pm 0.0654, which was by the state regulation.



Figure 3. Spatial and temporal variation of the pH

3.4 Electrical Conductivity (EC): Chemically pure water has low EC. The higher the EC of natural water, the more polluted it will be. In natural waters, mineralization and productivity reactions are similar. Natural water will have less or more mineral matter depending on when these two processes dominate. EC of water indicates

the general presence of chemical compounds and is an indicator of water pollution. This parameter in the four stations was different but not very pronounced. Thus, EC values ranged from 146.40 - 262.00 μ S/cm. The lowest value was measured in March in D3, while the highest value was in March in D1. The average values in March, April, May and June were 214.85; 218.52; 212.35 and 209.98 respectively (Fig. 4). The average seasonal value with standard deviation was 213.92 ± 3.6455 μ S/cm, which was within the allowed values of the state regulation.



Figure 4. Spatial and temporal variation of the EC

3.5 Chemical Oxygen Demand (COD): COD is usually used for the indirect measurement of the number of organic compounds in water. The main application of COD is to quantify organic pollutants found in surface water or wastewater, making COD a useful measure of water quality. COD is the amount of oxygen required to carry out the oxidation of organic pollution using a strong oxidizing agent. Research conducted on organic pollution of drinking water and liver cancer shows that mortality due to liver cancer is positively correlated with COD of drinking water. COD measurements have varied monthly with a range of 0,93 -1.82 mg/L. Thus, the lowest values were in D1 in June, while the highest value was in D2 in April, D3 and D4in March (Fig. 5). Average values during March, April, May, and June were 1.65; 1.52 1.35, and 1.24 mg/L respectively. The average seasonal value with standard deviation was 1.425 ± 0.1936 mg/L, which is within the allowed values of the state regulation.



Figure 5. Spatial and temporat variation of the COD

3.6 Total Dissolved Solids (TDS): TDS is the term applied to the residue remaining in a mass measuring vessel after the sample has passed through a standard glass fiber filter and dried to constant mass at 103 - 105 °C or 179 - 181°C. Water with high TDS content often has a laxative effect and sometimes the opposite effect on individuals whose bodies are not adapted to it. TDS mainly consists of Ca2+, Mg2+ ions, bicarbonates, carbonates, sulfates, chlorides, nitrates, and other substances. A high concentration of TDS around 3000 mg/L can also produce disturbance in animals. This parameter in four stations was different but not very pronounced. Thus, the values were brought from 315.00 - 390.00 mg/L. The lowest value was measured in March in D4, while the highest value was in May in D3 (Fig. 6). The average values during March, April, May, and June were 332.25; 350.75; 364.25 and 379.53 mg/L respectively. The average seasonal value with standard deviation was 356.69 ± 20.08 mg/L, which was lower than the recommended value of the state regulation.



Figure6. Spatial and temporal variation of the TDS

3.7 Chlorides: Chlorides are less dangerous contaminants in drinking water. According to the permitted standards, their content in river waters is quite high. Chloride ions occur naturally in surface and groundwater. They are also found in high concentrations in seawater. Higher than normal chloride concentrations in freshwater are detrimental to water quality. The use of road salt for winter accident prevention is a major source of chlorides for the environment. Unfortunately, the chloride content has increased over time due to road widening and increased ground traffic. The results of our measurements of chlorides in the drinking water of the city of Kumanovo for 4 months are shown in Fig. 7. Their concentration ranges from 5.12-6.62 mg/L. The lowest value was measured at D1, D2, D3, and D4 in June, while the highest value measured in May was at D1. The average values during March, April, May, and June were 5.93; 5.73; 6.43, and 5.14 mg/L respectively. The average seasonal value with standard deviation was 5.7938 ± 0.55 mg/L, which was within the allowed values of the state regulation.



Figure 7. Spatial and temporat variation of the chlorides

3.8 Calculation of THM content prediction in drinking water of the city of Debar by mathematical models: THMs prediction models are implemented based on routine measurements of some drinking water parameters and with their help, the concentration of THMs in drinking water is calculated. These predictive models contain different water quality parameters and individual models typically use three to eight parameters (Babaei *et al*, 2015). For the THMs prediction in the drinking water of Debar, we have used the equations of the ten mathematical models developed by (Durmishi, 2013). Mathematical model equations and the calculation of THMs content are given below. To obtain more reliable results for the prediction of THMs content for the spring season we have obtained the average value with a standard deviation of 23.88 \pm 8.16 µg/L (Table 1).

Calculation by:

Model 1:

THM = -10.925 + 0.688(WT) + 24.387(RC) + 0.461(pH) + 0.046(EC) + 2.076(COD) + 0.713(Chlorides) $THM = -10.925 + 0.688 \cdot (10.625) + 24.387 \cdot (0.2) + 0.461 \cdot (7.8225) + 0.046 \cdot (213.92) + 2.076 \cdot (1.425) + 0.713 \cdot (5.79375) = -10.925 + 7.31 + 4.8774 + 3.61 + 9.84 + 2.96 + 4.13 =$ **21.8 µg/L**

Model 2:

THM = 0.889 + 0.822(Chlorides) + 0.068(EC) + 21.205(RC) $THM = 0.889 + 0.822 \cdot (5.79375) + 0.068 \cdot (213.92) + 21.205 \cdot (0.2) =$ $= 0.889 + 4.76 + 14.55 + 4.241 = 24.44 \ \mu g/L$

Model 3:

log(THM) = 0.152 + 1.147 log(WT) + 0.158 log(RC) + 0.458 log(EC) - 0.557 log(TDS) + 0.252 log(Chlorides) + 0.240 log(pH) $log(THM) = 0.152 + 1.147 log(10.625) + 0.158 log(0.2) + 0.458 log(213.92) - 0.557 log(356.69) + 0.252 log(5.79375) + 0.240 log(7.8225) = = 0.152 + 1.147 \cdot (1.03) + 0.158 \cdot (-0.7) + 0.458 \cdot (2.33) - 0.557 \cdot (2.55) + 0.252 \cdot (0.76) + 0.240 \cdot (0.89) = 0.152 + 1.18 - 0.11 + 1.07 - 1.42 + 0.19 + 0.21 = 1.272 = 10^{1.272} =$ **18.71 µg/L**

Model 4:

 $THM = 1.419(WT)^{1.147} \cdot (RC)^{0.158} \cdot (PE)^{0.458} \cdot (Chlorides)^{0.252} \cdot (pH)^{0.240} \cdot (TDS)^{-0.557}$ THM=1.419 \cdot (10.625)^{1.147} \cdot (0.2)^{0.158} \cdot (213.92)^{0.458} \cdot (5.79375)^{0.252} \cdot (7.8225)^{0.240} \cdot (356.69)^{-0.557} = 1.419 \cdot 15.038 \cdot 0.775 \cdot 11.675 \cdot 1.557 \cdot 1.638 \cdot 0.038 = **18.71 µg/L**

Model 5:

log(THM) = 1.254 + 0.286 log(Chlorides)log(THM) = 1.254 + 0.286log(5.79375) = = 1.254 + 0.286 \cdot 0.763 = 1.254 + 0.218 = 1.472 = 10^{1.472} = **29.65 µg/L**

Model 6:

log(THM) = 0.113 + 0.256 log(Chlorides) + 1.31 log(WT)log(THM) = 0.113 + 0.256log(5.79375) + 1.31log(10.625) = =0.113 + 0.256 \cdot 0.763 + 1.31 \cdot 1.026 = 0.113 + 0.195 + 1.344 = 1.652 = 10^{1.652} = **44.87 µg/L**

Model 7:

log(THM) = 0.364 + 0.282 log(Chlorides) + 1.689 log(WT) - 0.372 log(TDS)log(THM) = 0.364 + 0.282log(5.79375) + 1.689log(10.625) - 0.372log(356.69) = = 0.364 + 0.282 \cdot 0.763 + 1.689 \cdot 1.026 - 0.372 \cdot 2.552 = 0.364 + 0.215 + 1.733 - 0.95 = 1.362 = 10^{1.362} = **23.01 µg/L**

Model 8:

log(THM) = 0.154 + 0.267 log(Chlorides) + 1.252 log(WT) - 0.532 log(TDS) + 0.431 log(EC)log(THM) = 0.154 + 0.267 log(5.79375) + 1.252 log(10.625) - 0.532 log(356.69) + 0.431 log(213.92) = 0.154 + 0.267 \cdot 0.763 + 1.252 \cdot 1.026 - 0.532 \cdot 2.552 + 0.431 \cdot 2.330 = = 0.154 + 0.204 + 1.284 - 1.358 + 1.004 = 1.288 = 10^{1.288} = **19.41 µg/L**

Model 9:

 $log(THM) = 0.340 + 0.258 log(Chlorides) + 1.030 log(WT) - 0.516 log(TDS) + 0.477 log(EC) + 0.153 log(RC) \\ log(THM) = 0.340 + 0.258 log(5.79375) + 1.030 log(10.625) - 0.516 log(356.69) + 0.477 log(213.92) + 0.153 log(0.2) = 0.340 + 0.258 \cdot 0.763 + 1.030 \cdot 1.026 - 0.516 \cdot 2.552 + 0.477 \cdot 2.330 + 0.153 \cdot (-0.7) = 0.340 + 0.197 + 1.057 - 1.317 + 1.111 - 0.107 = 1.281 = 10^{1.281} =$ **19.1 µg/L**

Model 10:

 $THM = 2.188 \ (Chlorides)^{0.258} \cdot (WT)^{1.030} \cdot (EC)^{0.477} \cdot (RC)^{0.153} \cdot (TDS)^{-0.516}$ THM=2.188 \cdot (5.79375)^{0.258} \cdot (10.625)^{1.030} \cdot (213.92)^{0.477} \cdot (0.2)^{0.153} \cdot (356.69)^{-0.516} = 2.188 \cdot 1.573 \cdot 11.406 \cdot 12.928 \cdot 0.782 \cdot 0.048 = **19.05 µg/L**

To obtain a more reliable result for predicting the content of THMs for the spring season, we obtained the average value with standard deviation of THMs, as in Table 1.

| Model | Values of THM content according to models |
|---------------------------|--|
| | (μg/L) |
| 1 | 21.8 |
| 2 | 24.44 |
| 3 | 18.71 |
| 4 | 18.71 |
| 5 | 29.65 |
| 6 | 44.87 |
| 7 | 23.01 |
| 8 | 19.41 |
| 9 | 19.1 |
| 10 | 19.05 |
| Average | 23.875 |
| Standard Deviation | 8.1574 |
| Content of THMs | $23.88 \pm 8.16 \ \mu\text{g/L}$ |

Table 1. Calculation of the mean value with standard deviation of THMs of ten models

4. Conclusion

From the results obtained during the analysis of the physicochemical parameters, namely the prediction of THMs in the city of Debar, we can conclude that:

• The parameter values were in accordance with the recommended values of the State Regulation on the quality of drinking water;

• According to the content of inorganic and organic substances, the drinking water of the city of Debar is of good quality and can be used for drinking;

• The models used for the prediction of THMs have given successful results;

• The recommended value for THMs according to the National and European Regulation is 100 µg/L;

• The prediction of THMs content with the 10 models for the spring season of 2021 was 23.88 ±

8.16µg/L;

- So, for the research period THMs do not pose a risk to the health of the population;
- THMs reduction should be encouraged, but without compromising drinking water disinfection; and
- We recommend the relevant authorities to take preventive steps to keep THMs under control.

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