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# Disinfection of Drinking Water and Trihalomethanes

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**ABSTRACT:** Best known by-products of chlorination of drinking water are trihalomethanes (THMs) which can have negative effects on human health. The research goals in this paper were to emphasise the importance of THMs as by-products of disinfecting drinking water, to show the practice of controlling the THMs in drinking water on the example of three water supply systems in the Republic of Srpska and to propose measures for reducing formation of THMs in drinking water. The results have shown that values of THMs in drinking water in these three water supply systems were within the reference values, but the level of the proscribed control is significantly below the minimal requirements.

**Keywords:** disinfection of drinking water, trihalometanes.

## INTRODUCTION

Disinfection is definitely the most important step in preparing water for public supply, but besides being strong biocides, disinfectants are also reacting with other components in water thus creating new compounds with potentially harmful long-term health side-effects. Therefore, a general assessment of the impact of disinfection on public health must take into account not only the microbiological quality of the treated water, but also the toxicity of disinfectants and their reaction products, or as they are commonly called, water disinfection by-products (Dalmacija at al., 2006.).

The type of disinfection by-products depends primarily on the applied disinfectant, and then on water quality parameters (type and concentration of natural organic matter, presence or absence of bromide and iodide ions, pH, temperature, organic nitrogen concentration, etc.), as well as on operating conditions (disinfectant doses and contact times) applied in order to simultaneously achieve effective disinfection and provide requirements for achieving residual disinfectant concentrations (Tričković at al., 2005.).

In our country, but also worldwide, the most commonly used disinfectant for drinking water are chlorine and chlorine preparations. The advantage of using chlorine for water disinfection is that it has rapid effect on microorganisms, it has residual effect which ensures safety of subsequent water pollution, and is an affordable method.

The best-known by-products of water chlorination are certainly trihalomethanes (THMs), out of which the following are important as water contaminants: bromoform, dibromochloromethane, bromodichloromethane and chloroform. Reducing the concentration of these four most commonly reported THMs should decrease the concentration of other uncharacterized chlorination by-products (Ivančev-Tumbas, 1998.). Chloroform is usually the most abundant in drinking water. Its effects on health are also most researched.

As early as the 1970s, research began on the possible effects of by-products resulting from the disinfection of drinking water on human health (Bellar at al., 1974; Hrudey, 2009; Rook, 1974.). In 1976, the National Cancer Institute published the carcinogenic effect of chloroform using rodent research (Boorman at al., 1999; Singer, 1993.). Following these results, in 1991 the International Agency for Research on Cancer of the World Health Organization classified chloroform and other widespread by-products of disinfecting drinking water as possible carcinogens (ARC, 1999). Looking at THMs as a whole, numerous studies examining the effect of THMs on human health have found that eating and drinking, transdermally or by inhalation can lead to a number of health issues, so THMs were classified as carcinogenic and mutagenic compounds (WHO; Lee at al., 2004; Min at al., 2016; Hang at al., 2016.).

Various meta-analyses and pooled studies in Europe and North America have provided consistent evidence that long-term exposure to t THMs is associated with an increased risk of bladder cancer and colorectal cancer (Kargalioglu at al., 2002; Gruau, 2004; Monarca at al., 2004; Costet at al., 2011; King at al 2006); Villanueva at al., 2003.). One recent multicentre study found no clear evidence of an association between total lifetime THMs exposure and colon cancer (Villanueva at al., 2017). If the activity of individual fractions was observed, then this study revealed a protective effect of chloroform, while a positive association with brominated trihalomethanes was observed in men at the tail end of the exposure distribution, with the key conclusion being that these results require confirmation. Other studies have also reported an association between these compounds and adverse reproductive effects such as congenital anomalies, cancer, and fetal development during pregnancy (Nieuwenhuijsen at al., 2009 A; Nieuwenhuijsen Nieuwenhuijsen at al., 2009 B.).

In studies of the human population using chlorinated drinking water in which chloroform is predominantly THMs, small increases in the incidence of rectal, colon, and bladder cancers have been consistently observed, and the strongest evidence was given for bladder cancer (Kargalioglu at al., 2002; Gruau, 2004; Monarca at al., 2004.). However, since other possible carcinogens have been found in this water, it is impossible to identify chloroform as the only carcinogen. Therefore, the U.S. Environmental Protection Agency classified chloroform as group B2 or “probable carcinogen for humans,” based on sufficient animal evidence and inadequate human evidence of carcinogenicity. Evidence from animal studies now strongly suggests that exposure to chloroform causes cancer only after it produces permanent cellular toxicity. Because a certain threshold level of exposure is required to induce cell toxicity, the occurrence of cancer due to exposure to chloroform is only possible if this threshold is exceeded.

In many countries, including the Republic of Srpska, the obligation to control trihalomethanes in drinking water is legally proscribed.

This study aimed to indicate the importance of determining THMs as a by-product of disinfecting drinking, to show the practice of controlling the THMs in drinking water on the example of three water supply systems in the Republic of Srpska and to propose measures for reducing formation of THMs in drinking water.

## MATERIALS AND METHODS

The research was conducted as a retrospective study that included the results of analysis of drinking water samples from the central water supply systems of Doboj, Derventa and Teslić on total THMs and its main fractions, in the period 2017 - 2019. The Central Protocol of the Laboratory of the Public Health Institute of the Republic of Srpska - Banja Luka was used as data source.

Water sampling was performed by experts from the Institute of Public Health of the Republic of Srpska in accordance with Water quality – Sampling – Part 3 : Preservationand handling of water samples( BAS EN ISO 5667-3.2018) and Water quality – Sampling – Part 5: Guidance on sampling of drinking water from tretment works and piped distribution systems ( ISO 5667 -5). The analyzes were performed in the Laboratory of Sanitary Chemistry of the Institute of Public Health of the Republic of Srpska, by the method of gas chromatography with the use of ECD detectors.

Determination of the correctness of water samples was performed according to the Rulebook on the health safety of water intended for human consumption [23]. The results of the analyses are presented in the form of tables. Descriptive statistics methods were used in data processing.

## RESULTS

In the observed period 2017-2019 it has been found that in the city water supply system in Doboj, the analysis for the presence of total THMs was performed within the regular basic water inspections, performing sampling once per month (Table 1)

**Table 1.** Analyses results of water samples from the central water supply system in Doboj for total trihalomethanes in the period 2017 – 2019 (µg/L)

| Month | 2017 | 2018 | 2019 | Reference value (µg/L) |
|---|---|---|---|---|
| January | 5,3 | <0,5 | <0,5 | ≤100 |
| February | 2,4 | <0,5 | 4,5 | ≤100 |
| March | 2,3 | <0,5 | 11,2 | ≤100 |
| April | 3,5 | 1,8 | 23,0 | ≤100 |
| May | 2,2 | | 7,6 | ≤100 |
| June | 3,0 | 0,8 | 4,4 | ≤100 |
| July | 5,3 | <0,5 | 18,9 | ≤100 |
| August | 3,0 | 6,0 | 4,1 | ≤100 |
| September | <0,5 | 0,5 | 8,6 | ≤100 |
| October | 2,6 | 1,0 | 14,4 | ≤100 |
| November | 2,9 | 3,6 | 13,2 | ≤100 |
| December | 1,7 | 12,6 | 4,8 | ≤100 |

All values of total THMs were within legally permitted concentrations (≤100 µg/L) (Službenik glasnik Republike Srpske 88/17). The highest concentration of total THMs was recorded in April 2019 being up to 23.0 µ g / L, which is four times less than the maximum allowed concentration.

Although these are acceptable concentrations, there is a noticeable increase in the concentrations of total THMs in 2019 compared to the previous two years.

From June 2017, the analysis of drinking water for certain fractions of THMs was started: chloroform, bromodichloromethane, dibromochloromethane, bromoform. Since chloroform is of the greatest importance, these are the results shown in Table 2. As in the case of total THMs, all chloroform concentrations were within the permitted values. The highest concentrations of chloroform were recorded in April and July 2019, and in December 2018.

**Table 2.** Analyses results of water samples from the central water supply system in Doboj for the presence of chloroform in the period 2017 – 2019 (µg/L)

| Month | 2017 | 2018 | 2019 | Reference value (µg/L) |
|---|---|---|---|---|
| January | - | <0,5 | <0,5 | ≤100 |
| February | - | <0,5 | 1,2 | ≤100 |
| March | - | <0,5 | 9,2 | ≤100 |

| | | | | |
|---|---|---|---|---|
| April | - | 0,5 | 19,8 | ≤100 |
| May | - | - | 4,0 | ≤100 |
| June | - | <0,5 | 2,2 | ≤100 |
| July | 1,2 | <0,5 | 13,0 | ≤100 |
| August | <0,5 | <0,5 | 1,8 | ≤100 |
| September | <0,5 | 0,5 | 3,6 | ≤100 |
| October | <0,5 | <0,5 | 9,5 | ≤100 |
| November | <0,5 | 0,8 | 9,5 | ≤100 |
| December | - | 11,7 | 1,2 | ≤100 |

The water analysis of the water supply system in Teslić was performed as part of the periodic water inspection in September 2017 and November 2018, and the obtained results are shown in Table 3.

**Table 3.** Analyses results of water samples from the central water supply system in Teslić for the presence of total trihalomethanes in 2017 and 2018 (µg/L)

| Parameter | 2017 | 2018 | Reference value (µg/L) |
|---|---|---|---|
| Trihalomethanes - total | 22,4 | 15,0 | ≤100 |
| Chloroform | 21,4 | 14,0 | ≤100 |
| Bromodichloromethane | 1,0 | 1 | ≤100 |
| Dibromochloromethane | <0,5 | <0,5 | ≤100 |
| Bromoform | <0,5 | <0,5 | ≤100 |

Concentrations of total THMs and individual fractions were within the prescribed limits.

**Table 4.** Analyses results of water samples from the central water supply system in Derventa for the presence of total trihalomethanes in 2017(µg/L)

| Parameter | 2017 | Reference value (µg/L) |
|---|---|---|
| Trihalomethanes - total | 0,9 | ≤100 |
| Chloroform | <0,5 | ≤100 |
| Bromodichloromethane | <0,5 | ≤100 |
| Dibromochloromethane | 0,9 | ≤100 |
| Bromoform | <0,5 | ≤100 |

The control of the presence of THMs in the water from the Central Water Supply of the Municipality of Derventa was performed once a year, as part of the periodic inspection, only in 2017. The obtained results of total and individual THMs in 2017 are shown in Table 4.

## DISCUSSION

In the Republic of Srpska, according to the Rulebook on the health safety of water intended for human consumption, total THMs should be determined as an integral part of the basic inspection of drinking water (Službenik glasnik Republike Srpske 88/17).

In Teslić and Derventa, the control was performed only in 2017 and 2018, i.e. 2017, namely the analysis of one sample within the periodic inspection of water. So it can be said that the minimal control that has just been started, has been stopped.

Determination of THMs in drinking water from the central water supply system in Doboj was performed in the observed period of three years doing one analysis per month. However, if we take into account that the basic water inspection in Doboj is performed three times a month, when each time sampling and analysis of water is performed with seven or eight control points, then the stated number of analyzes for THMs is far below the prescribed norms.

The maximum permissible concentration of total THMs in drinking water in the Republic of Srpska is 100 μg / l, 23 which is in accordance with the recommendations of the World Health Organization, i.e. the European Union regulations on maximum permissible pollution of drinking water with total THMs which is the sum of chloroform, bromodichloromethane, dibromochloromethane and bromoform (European Commission, 1998.).

The results of analyses of drinking water from the central water supply system in Doboj for THMs showed that all values of THMs in the observed period were within the allowed limits. The highest values of total THMs s were recorded in April (23.0 μg / l) and July (18.9 μg / l) 2019, which was again four to five times lower than the maximum permitted concentrations for total THMs. The highest measured concentrations of THMs in water samples were recorded mostly in the summer months, while the lowest values were in the first months of the observed years. Such variation in THMs concentrations can be related, in particular, to a change in the amount of organic matter present in the water. The upward trend during the summer months compared to the downward trend in the colder period of the year is characteristic of temperate climates (Gray, 2008; Brown at al., 2011.). Temperature and seasonal variation of natural organic matter also affect the formation of THMs, while during summer months, due to rinsing of organic matter, an increase in the formation of THMs was observed compared to the winter period. If we analyse the values of total THMs by years, then it is clear that the highest concentrations were recorded in 2019.

As already presented in the results, from June 2017 the concentration values for individual THMs (chloroform, bromodichloromethane, dibromochloromethane and bromoform) were also examined. Chloroform concentrations have shown that the values of total THMs largely “depend” on the values of chloroform. This means that the values of chloroform, as with total THMs, are highest in the summer months and lowest during low temperature periods.

Bromodichloromethane, dibromochloromethane and bromoform are three compounds with bromine that are also formed as by-products of water disinfection, due to the presence of organic matter, but are primarily formed with increased bromide content, which is not present in large quantities in fresh natural waters (Dogančić, at al.) Thus, in the presence of bromide, brominated trihalomethanes are formed, while the concentration of chloroform decreases proportionally.

Given the extremely low, extremely negligible number of analyses of drinking water from the water supply system in Derventa and Teslić for trihalomethanes, we cannot give any comments on those results.

The problem of THMs has so far been insufficiently dealt with by the profession and science in the Republic of Srpska. Sufficient proof for this claim is the fact that even in the annual reports of drinking water analyses of the Public Health Institute of the Republic of Srpska (authorized by law for public control of trihalomethanes in drinking water) the analysis of trihalomethane has not been given a single word.

At the same time, from the Report on the health safety of water for human consumption in the Republic of Croatia for 2018 (Hrvatski zavod za javno zdravstvo), published by the Croatian Institute of Public Health, we find out that 584 analyses for THMs were performed in the observed year, and that only in the case of one analysis (0.2%) the value of THMs exceeded the legally allowed maximum value of 100 μg / l.

Epidemiological studies have long been conducted in various European countries indicating great variability in levels within (Villanueva at al., 2017.) and between (Jeong at al., 2012.) the countries. European research project Health impacts of long-term exposure to disinfection by-products in drinking water (HIWATE), reported that in 2010 levels of THMs in drinking water in seven cities in five European countries ranged from below the detection limit (Modena, Italy) to above the currently regulated maximum limit (Barcelona, Spain) (Jeong at al., 2012.).

The situation in the countries of the European Union, and the importance of determining trihalomethanes and interpretation of the obtained findings, and if necessary taking certain measures, were best shown by the results of the European project EXPOsOMICS whose goal was to calculate the estimate of THMs levels in drinking water all over the Europe and assess the associated burden of bladder cancer using different exposure scenarios. This study included 28 countries of the European Union. The findings showed that current average levels of THMs in drinking water in all EU countries were below European regulatory limits, although maximum levels showed exceedances in nine countries. The key conclusion of this study was that assuming a cause-and-effect relationship, current exposure to THMs in the European Union could lead to a significant number of bladder cancer cases that could be avoided by optimizing water treatment, disinfection and distribution, among other measures, without compromising microbiological quality of drinking water (Evlampidou at al). [30]

The amount of THMs that will be formed depends on the amount of precursors, the amount of chlorine used for disinfection and the length of contact between water and chlorine (longer contact means higher production of THMs) (Matošić at al., 2007.). There are several options for controlling disinfection and disinfection by-products: moving the disinfection point, removing by-products when they have already been created, removing precursors or natural organic matter before reacting with disinfectants, or using disinfectants that minimize by-product formation.

The cheapest approach to control is to move the disinfection point or use an alternative disinfectant. The least desirable approach is to remove disinfectant by-products when they have already been created.

The best approach to control is to remove precursors before they react with the disinfectant. Currently, a large number of different techniques are used to reduce the content of by-products precursors in water, of which the most commonly used are conventional physicochemical methods, such as coagulation processes with iron and aluminium salts, filtration on single-medium and dual-media filters, and GAU filtration (Yan at al., 2007).

## CONCLUSION

Trihalomethanes (chloroform, bromodichloromethane, dibromochloromethane, bromoform) are the most important by-products of water disinfection by chlorine and chlorine compounds.

Previous studies have shown that THMs are probable or possible carcinogens, that they can have a mutagenic effect, and that they affect the occurrence of respiratory diseases.

Due to risk to human health, determination of THMs in drinking water, which is still predominantly being chlorine disinfected, is done.

In the period 2017-2019, the analysis of drinking water for total THMs, as well as for certain compounds (chloroform, bromodichloromethane, dibromochloromethane, bromoform) in drinking water from Doboj water supply system was performed once a month.

In the period 2017-2019, in Teslić and Derventa, the control was performed once a year, as part of the periodic inspection of drinking water, but not on a regular basis (analyzes in Teslić were done in 2017 and 2018, in Derventa only in 2017).

Water control level in Doboj water supply system, and especially in the water supply systems in Derventa and Teslić, is below the proscribed level.

The results of the analysis for THMs (chloroform, bromodichloromethane, dibromochloromethane, bromoform) in water samples from Doboj water supply system showed that the obtained concentrations THMs (individually or in total) were not above the permitted value (MDK 100 µg / L).

The highest measured concentrations of trihalomethane were recorded mostly during summer months, while the lowest values were in the first months of the observed years, which is most likely related to the change in the amount of organic matter present in the water, which is characteristic of temperate regions.

Although still significantly below the maximum allowable concentrations for total THMs and compounds, there is an evident increase in their concentrations in 2019 compared to the previous two years.

Measures to reduce the formation of THMs in drinking water are the removal of precursors or natural organic matter before they react with disinfectants, the use of alternative disinfectants that minimize the formation of by-products, and the least desirable approach is the removal of disinfectant by-products when already created.

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