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# TRIHALOMETHANES REMOVAL FROM DRINKING WATER BY HEATING: A CASE STUDY

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## ABSTRACT

Trihalomethanes (THMs) specially Chloroform is the most significant and well-known by-products of chlorine disinfection. In light of the alleged carcinogenic effects of THMs in drinking water, the goal of this study was to investigate the consequences of heating the chlorinated water, to lower its THM content, if any, in the drinking water of Dhaka. In this experiment, three individual samples from three separate locations in the same water supply system were investigated in 2019. Chloroform concentrations decreased noticeably (from 42% to 100%) due to heating during the trials. The results revealed that in sample 1, chloroform decreased from 199 to 79.7 20 µg/l (60% removal) after one minute of boiling of water, to 20 µg/l (90% elimination) after two minutes, and finally to 0 µg/l (100% removal) after three minutes. The comparable reductions in samples 2 and 3 were also similar: 70 to 24.5 µg/l (65% removal), 8.5 µg/l (88% removal), and zero µg/l (100% removal) and 153 to 89 µg/l (41.83% removal), 41 µg/l (73.2% removal), and to 40 µg/l (73.86% removal). According to experimental findings, if THM precursors and chlorine residuals are present, there may be a further considerable synthesis of THMs during the heating of drinking water. The generation and volatilization rates of THMs were responsible for the overall fluctuation in THM content in water during heating. Temperature and reaction time were major determinants of the rate of THMs synthesis, in addition to the amounts of THMs precursors and residual chlorine.

**Keywords:** Chlorination, Disinfection By-Products, Drinking Water, Heating, Trihalomethanes

## 1. INTRODUCTION

For humanity, having access to drinking water that meets acceptable standards is a top priority. The standard of a country's drinking water determines its intellectual ability, and a civilization's advancement is based on the sophistication of its water supply and sewage systems. Water is crucial to human pathology since sources of many diseases are transmitted by water. These may come from the improper selection of a water treatment procedure that may not adhere to the fundamental hygienic standards, sanitation practices, and disinfection in particular. Bujar et al. (2015).

Pathogens, toxic chemicals, and odour-causing pollutants are all things that drinking water treatment aims to get rid of from raw water. A standard water treatment procedure is frequently utilized for surface water sources, such as rivers, lakes, and reservoirs. Coagulation, flocculation, sedimentation, filtering, and disinfection are all parts of the process. While flocculation turns small particles into large settleable or filterable flocs, coagulation balances the charge on the surface of particles. These flocs and other neutralized particles, including pathogens, are eliminated through sedimentation and filtration. Disinfection is the last step and has two main goals. Pathogens are primarily eliminated or rendered inactive by disinfection. Providing a residual disinfectant in finished water and preventing microbial regrowth in water distribution systems are the secondary goals. Although coagulation, flocculation, sedimentation, and filtration eliminate 99 to 99.9% of germs, disinfection is still a vital step in protecting the public from developing waterborne illnesses. Yuefeng (2005), Barrett et al. (2000).

Chlorination has been used to disinfect drinking water for more than a century and it has eliminated the majority of waterborne diseases. According to the American Water Works Association (AWWA), chlorine is used as a disinfectant in more than 80% of water treatment facilities in the USA AWWA (2000). Similar to this, the majority of water treatment facilities in Canada also utilize chlorination to disinfect water Health Canada (2006). Chlorine is the only disinfectant used in all public water treatment facilities in Bangladesh.

Chlorine is a highly powerful disinfectant against waterborne microbiological agents and offers prolonged protection to prevent microbial growth in water distribution systems ACC (American Chemistry Council) (2018). Chlorine is still the most efficient and affordable disinfectant, despite the availability of various disinfectants and disinfection techniques (chlorination, chloramination, chlorine dioxide, granular activated carbon with post-chlorination, ozonation, and ultraviolet radiation) Clark et al. (1998), Reiff (1995), Chowdhury et al. (2007). The increased oxidizing potential of chlorine, which ensures a constant level of chlorine residual throughout the distribution system and prevents microbial recontamination, is another factor contributing to its appeal in addition to its lower cost.

**The Challenge of Disinfection By products**

Drinking water systems must control disinfection by products (DBPs), chemical compounds formed unintentionally when oxidants like chlorine and other disinfectants react with naturally occurring organic matter in the source water. While acute microbial contamination protection is of the utmost importance, DBP control is also necessary Chowdhury et al. (2007). Trihalomethanes (THMs), a family of DBPs that includes chloroform, were found to be produced by drinking water chlorination in 1974, according to separate findings by EPA scientists and a Dutch researcher Rook (1976).

Three hydrogen atoms in methane are replaced with halogen atoms to produce THMs. Only four THMs—chloroform ($CHCl_3$), bromodichloromethane ($CHBrCl_2$), dibromochoromethane ($CHBr_2Cl$), and bromoform ($CHBr_3$)—are the principal ones produced during the chlorination process. Total trihalomethanes are all four taken collectively (TTHMs). In 1979, the EPA established the first THM regulating levels. Despite the fact that the body of studies does not conclusively demonstrate that high concentrations of disinfection byproducts (DBPs) in drinking water have a negative impact on human health, their presence is undesirable. Chloroform is always present in the highest proportion in drinking water, and it frequently makes up more than 90% of the overall concentration of THMs Bhujar (2013), Chang et al. (2001), Chan (1997). In non-coastal zones, where bromoform is typically absent, a study showed that chloroform contributed the most to the total amount of THMs (97.99 to 98.71%). Mishra (2016).

The concentration of THMs during the distribution process is influenced by a number of variables, such as contact times, water flow pathways, system maintenance conditions, and residual disinfectant maintenance procedures. The increase in the concentration of THMs is supported by the high values of factors including organic matter, temperature, pH, chlorine dosage, bromides, and contact times Chang et al. (2008), El-Shahat et al. (2001). THMs have received more attention in recent years in the USA and Europe, whereas they have received less attention in Southeast Asia and developing nations because most water authorities there are comfortable with the standard of World Health Organization (WHO) recommendations of 1984.

Changes in process conditions (removal of precursor chemicals), the use of various chemical disinfectants, or the use of non-chemical disinfectants (UV radiation or the membrane process) are the three main methods used to reduce the concentration of THMs. The 1998 European Council Directive for drinking water sets a limit on THMs of 100 g/L started in 2008. EU (2020). The level of THMs in drinking water is being reduced as new standards for regulating and managing them are continuously adjusted. While some nations have established the maximum limit for THMs, others have not yet done so.

Heating is one of the techniques for removing THMs After these compounds are formed Ebrahim et al. (2016). Each THM is eliminated using this procedure at a particular boiling point, which is based on the chemical process of evaporation Wells et al. (2001). The traditional methods for removing these compounds, such as the use of activated carbon fibers Uchid et al. (1997), granular activated carbon Koumenides et al. (2001), membrane processes Vedat et al. (2008), ultrasonic methods Khordehdan et al. (2014), Air Stripping Packed - Column Samadi et al. (2006), etc., require expensive equipment, chemicals, and certain laboratory conditions (pilot scale, etc.). Therefore, the heating process's applicability can be increased by using realistic, affordable procedures that don't include chemicals and have a high removal efficiency.

The majority of studies on DBP generation have been carried out between 5 and 300 °C. But before being consumed (for example, when cooking or brewing tea or coffee), tap water is frequently heated or boiled. It is also heated for a number of other household functions (showering, laundry, etc.). Even a large number of individuals in third-world nations regularly consume water that has been boiled and then cooled. According to recent research BIGD (2019) in Dhaka, 42 percent of the city's residents boiled the water the city authority supplied before drinking it. Such heating could remove volatile DBPs from the solution, produce more and possibly different DBPs through interactions with residual disinfectant and natural organic matter (NOM), and change current DBPs into new DBPs and/or non-DBP compounds Weisel and Chen (1994). Although a few researchWeisel and Chen (1994), Kim (1997), Dojlido et al. (1999), Liu (2014), Batterman et al. (2013), Yamamoto et al. (2005) examined the generation and behaviour of DBPs in hot chlorinated tap water, the data are scarce and do not cover all of the frequently reported DBPs.

**Objectives of the study**

Analysis of samples of raw water from the drinking water source in Dhaka revealed the presence of organic pollutants at a higher level than usual, particularly in the dry season. This study was intended to investigate the effects of heating the water on the elimination or reduction of the amount of THMs in drinking water in the city, to expand that database, and to see if the people of Dhaka who use to drink the city water supply are receiving any additional benefits by boiling the supplied water.

## 2. MATERIALS AND METHODS

At present twenty-two percent of total potable water in Dhaka, the capital city of Bangladesh having fifteen million denizens, is treated from Sitalakhya river water. In case of inadequate and improper treatment techniques, there is a likely chance of occurrence of THMs in the water supplied. The largest drinking Water Treatment Plant in Dhaka which draws raw water from the Sitalakhya river was established and put into operation in 2002. The largest drinking water treatment plant beside the Sitalakhya is equipped with clarifiers consisting of coagulation & flocculation facilities and preceded by a biological pretreatment unit. To carry out this investigation three individual water samples from three different locations of the drinking water supply system were collected.:

1) On 22 December 2019 water samples were collected from a tap in the southeast part of Dhaka from a treated water supply in an area adjacent to the treatment plant. This water most likely was a mixture of surface water and groundwater supply, however, the actual mixing ratio was unknown. The tap water was allowed to run for at least 10 min in order to stabilize the temperature and residual chlorine concentration of the water. Some of the relevant water pollution parameters of the samples are given in Table 1. Chlorine was added to the samples at a concentration so that the initial resulting THM concentration stands a bit higher, and the initial free residual chlorine residual concentration stands at a figure of more than 0.9 mg /L.
2) On 07 November 2019, pretreated water samples from the treatment plant were collected which water quality characteristics are shown in Table 1. Chlorine was added to the samples to make the pretreated water chlorinated and allowed for a contact time of 6.5 hours to get a visible initial resulted THM concentration. The resulting water quality parameters after 6.5 hours are shown in Table 2.
3) On 09 November 2020, treated water samples from the treatment plant were collected and water quality characteristics are also shown in Table 1.

**Table 1**

**Table 1 Status of Water Quality Characteristics of the Samples Under Test**

| Sample ID and date of sampling | pH | Turbidity | $NH_3$-N | Free $cl_2$ | Total $Cl_2$ | COD | DOC | $UV_{254}$ | Conductivity | TDS | DO | T° C |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Pre-treated Water (7/11.19) | 7.13 | 37.1 | 0.33 | 0 | 0 | 14 | 7.2 | 0.1012 | 221 | 143.9 | 7.63 | 27.5 |
| treated Water (9/11/20) | 6.85 | 0.22 | 00 | 1.22 | 1.63 | 8 | - | 0.045 | 246 | 126 | 7.53 | 29.6 |
| Tap Water (22/12/19) | 7.08 | 0.36 | 0.0 | 0.89 | 1.29 | 11 | 6.0 | 0.0616 | 386 | 251 | 9.06 | 23.0 |

A 1000 ml serialized container made of dark glass was used for water sampling, The container before sterilizing was washed multiple times with detergent and diluted hydrochloric acid. Two samples were taken from each sampling at the same time. The first sample was kept intervened. The second sample was heated after pouring in a 2000 ml beaker on a laboratory digital hotplate for boiling. After 1, 2, and 3 minutes of boiling Three samples from each category of the samples under investigation were taken from the heated boiled and kept in a sampling container and stored in presence of ice, and within twelve hours taken to and used in the laboratory of the largest treatment plant equipped with advanced equipment. After cooling the samples were analysed according to Standard Methods. Chloroform was measured by THM plus Method (Method:10132) using UV-VIS Spectrophotometer DR 6000(HACH, USA).

**Table 2**

**Table 2 THM Analysis**

| | **Sample 01** | **Sample 02** | **Sample 03** |
|---|---|---|---|
| **Sample** | **Pre-treated Water (7/11/19)** | **treated Water (9/11/20)** | **Tap Water (22/12/19)** |
| Date of testing | November 07, 2019 | November 09,2020 | December 22, 2019 |
| Chlorine added | 6.5 | 0.0 | 6.82 |
| Ammonia added | 0.0 | 0.0 | 0 |
| **After 6.5 Hours of chlorine addition** | | | |
| pH | 7.18 | 6.85 | 7.26 |
| Turbidity | 45.9 | 0.22 | 0.36 |
| Ammonia-N | 0.0 | 0.00 | 0.03 |
| Free Chlorine (mg/L) | 1.17 | 1.22 | 0.35 |
| Total Chlorine (mg/l) | 1.62 | 1.63 | 0.85 |
| Mono-chloramine | 0.0 | NA | 0.0 |
| COD (mg/l) | 7 | 8 | 10 |
| DOC (mg/l) | 9.4 | - | 5.8 |
| UV 254 nm | - | 0.045 | 0.0556 |
| THM ($CHCl_3$) (ppb) | 199 | 111 | 153 |
| Conductivity (µS/cm) | - | 246 | 380 |
| TDS (mg/L) | - | 126 | 247 |
| DO (mg/L) | - | 7.53 | 8.93 |
| Temperature | - | *29.6* | *20.3* |

## 3. RESULTS AND DISCUSSIONS

In this study, the concentration of chloroform was determined, and it was assumed that that the total concentration of trihalomethanes (TTHMs) would be a bit higher assuming that chloroform is 95% of the total THMs in any sample under the study Mishra (2016). In this paper, we use THM and chloroform interchangeably. The initial concentrations of all trihalomethanes before heating in three water samples under study are shown in Table 3. The sample 2, (treated water), the THM concentration is less than the standard set by the Bangladesh National Standards (ECR, 1997) (90 µg/l for chloroform and World Health Organization (100 µg/l for all the trihalomethanes and EPA (80 µg/l for trihalomethanes. Sample 1 (Pretreated water) and sample 3 (Tap water) had concentrations of chloroform exceeding the above-mentioned standard limit. It is to be noted that in sample 3 the original tap water was added with additional chlorine in the tap water before heating to get THM at least somewhat higher than the allowable concentration set by Standards. However, in the raw sample from the system, that is in the tap water the concentration was a little less than 80 µg/l, the maximum concentration allowed by USEPA. In sample 3 also chlorine was added adequately to get the initial THM value at the higher side.

Table 3 and Figure 1 show the initial & the final concentration of trihalomethanes before and after heating/boiling as well as the changes in the tested samples No.1, 2 and 3. This shows that boiling distinctly reduces the THM concentrations in all samples.

In the case of sample 1chloroform reduced from 199 to 79.7 µg/l that is 60% THMs was removed. After 2 minutes of boiling chloroform reduced from 199 to 20 µg/l that is 90% THM was eliminated. The increase in removal from the first minute to the second minute is 30%. After 3 minutes of boiling the removal was 100%, from 199 to 00µg/l that is 10% removal between the second and third minute.

Boiling sample no. 2 for 01 min removed 65 % of all the chloroform that is chloroform reduced from 70 to 24.5 µg/l from the water. After 2 minutes of boiling chloroform reduced from 70 to 8.5 µg/l that is 88 % THM was eliminated. The increase in removal from the first minute to the second minute is 21%. After 3 minutes of boiling the removal was 100%, from 70 to 0 µg/l that is 12% removal between the second and third minute.

Boiling sample no. 3 for 01 min removed 41.83 % of all the THMs that is chloroform reduced from 153to 89µg/l from the water. After 2 minutes of boiling chloroform reduced from 153 to 41 µg/l that is 73.20% THM was eliminated. The increase in removal from the first minute to the second minute is 31.37%.

After 3 minutes of boiling the removal was 73.86%, from 153 to 40 µg/l that is only 0.66% removal between the second and third minute.

Therefore, it can be concluded that when chlorinated water undergoes heating or boiling it leads to changes in DBP concentration due to chemical reactions & volatilization, both and when solutions are boiled THMS is eliminated almost completely. It resembles earlier studies Glòria et al. (2013), Stuart et al. (2005), Wells et al. (2001).

**Table 3**

**Table 3 Initial & Final Concentrations of Trihalomethanes (Chloroform) of the Tested Water Samples and Corresponding % Removal After Heating**

| **Sample number and identification** | **Initial concentrations of Chloroform(µg/l)** | **Chloroform Concentrations (µg/l) after boiling / (% removal)** | | |
|---|---|---|---|---|
| Time elapsed | 00 minute | 01 minute | 02 minutes | 03 minutes |
| 1. Pre-treated Water (7/11/19) | 199 | 79.7 (60%) | 20 (90%) | 0 (100%) |
| 2. Treated Water (9/11/20) | 70 | 24.5 (65%) | 8.5 (88%) | 0 (100%) |

| | | | | |
|---|---|---|---|---|
| 3. Tap Water (22/12/19) | 153 | 89 (41.83%) | 41 (73.2%) | 40 (73.86%) |

It was reported by Wells et al. (2001) that heating/boiling the water could be utilized as an operative technique to reduce the concentrations of DPB in water due to its volatility characteristics. This statement was a result of a study by Wells.

Nevertheless, it may also be said that a higher duration of heating the water may result in an increased volume of DPB removal, which resembles the study by Ebrahim et al. (2016) furthermore, once a reduction in the concentration of DPB in water made through heating their elimination would be executed a bit slow which is apparently contrary to the findings of Ebrahim. However, this lesser pace in THM reduction may happen due to the further formation of THM during heating of the water under test & summing up with the existing THM concentrations. As the elimination was higher than the formation so the net result was a reduction in the THM value. An earlier study also showed a similar outcome Li and Sun (2001).

The outcome of this study may be expounded & warranted when we review that chloroform has a boiling point of 61.2 °C. Chloroform constitutes the lion's share of THMS in Dhaka's water – with this fact, heating/boiling can be a suitable approach to remove THM from water during emergency needs since it has the minimum cost, need no laboratory utensils as well as is effective to remove this unsolicited chemical.

In earlier studies, it is found that the THM volatilization is mainly affected by the temperature of water containing THM and the notch of agitation of the same water. With the increase in temperature up to boiling and agitation THM volatilization increases Kuo et al. (1997).

**Figure 1**

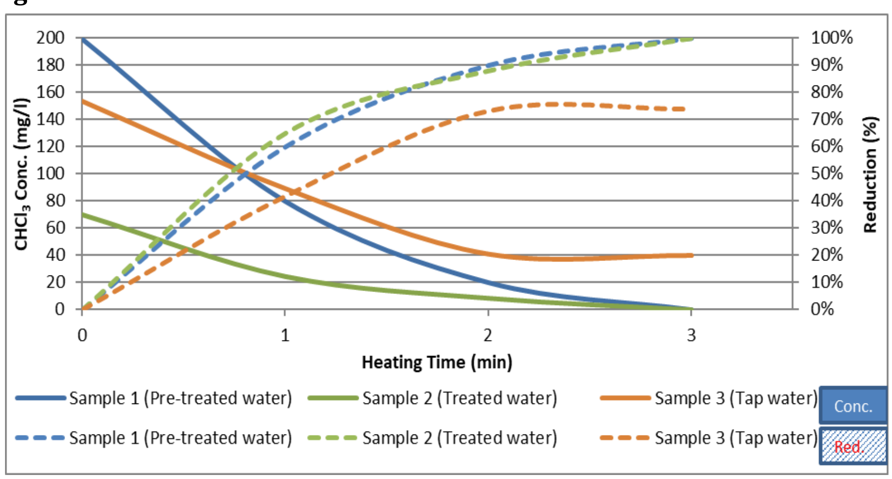


**Figure 1** Chloroform(mg/l) and its Percentage Removal (%) with Boiling Time(min.)

If anybody wants to eliminate THM from water by heating it is advised Wells et al. (2001) that water should undergo continuous heating beyond boiling for a few minutes. The presently available in market automatic domestic water heaters which are switched off automatically after boiling could be improved for THM minimization. The existing practice of using mostly gas ovens with continuous modes of burning as generally used in Dhaka is okay for this purpose.

The outcome of this study and that of Stewart et al. (2005) are similar. In the latter after 1,2 1nd 5 minutes of boiling the water the removal of chloroform and dichlorobromomethane was respectively 75%, 84%, and 97%; However, in our study boiling was made for 3 minutes not 5 minutes & 100% chloroform was removed in two cases out of three cases and in the third case removal was 74% after three minutes.

According to the results of their research Stewart et al. (2005), the initial concentrations of chloroform (569 μg/l) were reduced by 67.3% after 1 minute of boiling &82% after 5 minutes. However, in our study, the initial concentration of chloroform was relatively lower (199,153 μg/l) and it was expected to get a better removal percentage.

It can be noted that by comparing that in sample 1 though the initial concentration of THM was higher than that of sample 3, the reduction rate in sample 1 was faster than the sample 3 with respect of time and also in sample 1 the elimination was complete, which is not the case in sample 3. In this respect, if we examine the water quality parameters of the sample prior to heating we found that in sample 3 there is 0.89mg/L of free chlorine in sample 3, which is absent in sample 1. It can be concluded that due to the presence of free chlorine & some organic material in sample 3, during heating further THM was generated due to heating and was added with the initial concentration of THM, for this reason even after 3 minutes of boiling all the THM was not removed from sample 3, that is, volatilization & chemical reactions generating new THM was going simultaneously. That is thermal annihilation of chlorinated NOM can produce some THMs. However, the change in THM concentration due to heating is dominated by volatilization.

We know from earlier studies that if the water contains THM precursor & residual chlorine it could produce a further significant amount of THM during heating. Variation of THM concentration in water during heating depends on its rate of formation and volatilization. The rate of THM formation is affected by temperature & reaction time as well as the concentration of THM precursors & residual chlorine. Thus, we can conclude that the outcome of heating/boiling on DPBs are dependent on water temperature, type of residual disinfectant present, degree of DPB volatility & compound stability along with the quality of DPB precursors & chlorine residual present in the water.

As the raw water quality of the Sitalakhya river, particularly in dry months of the year, deteriorates notably, temperature increases, algae appear, and the water must be treated as it is designed, or it would increase trihalomethane concentrations in the drinking water. According to water officials of Dhaka, even today in the plant removal of DPB precursors is possible before chlorination,

but if the deterioration of the raw water quality goes on continuously by the vested interest group the future of water treatment is uncertain. It urges immediate actions to stop further raw water quality deterioration from THM precursors thus reducing the risks of city dwellers' exposure to DPB, via drinking water.

## CONFLICT OF INTERESTS

None.

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