Monitoring seasonal variations of haloacetic acids (HAAS) in low-TOC and low-chlorine networks and assessing risk to public health: Muş, Türkiye case

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Chlorine not only removes parasitic pathogenic microorganisms in water, but also causes the formation of disinfection by-products (DBPs) that can be carcinogenic to humans, due to reacting with natural organic matter (NOMs) in raw water sources. Haloacetic acids (HAAs) are one of the most well-known and common disinfection by-product groups (DBPs) in the literature. In the risk definitions of the EPA, some of its components have been identified as carcinogenic. Therefore, determination of HAA concentration in water and execution of a risk analysis are very important in terms of determining the possible effects on public health. This study aimed to monitor the seasonal and spatial variations of haloacetic acids (HAAs) in 2 different water supplies (surface and groundwater) serving the city center of Muş Province, Türkiye, and to demonstrate their public health implications. In this context, an analytical study was conducted covering 4 seasons. According to the results, although the amount of NOM in water bodies was less than $1~mg \cdot L^{-1}$, the HAA_5 content may occasionally exceed the USEPA limits of $60~\mu g \cdot L^{-1}$, but did not exceed the Canadian $80~\mu g \cdot L^{-1}$ limit. When the WHO limit values were examined on a component basis, it was determined that the MCAA concentrations in both water sources sometimes exceeded the limit of $20~\mu g \cdot L^{-1}$ in the July and October sampling periods. The risk level related to maximum DCAA level in the main network by means of ingestion pathway was found to be 18.7 times higher for women and 16.5 times higher for men when compared with USEPA risk criteria. Also, in the Muratpaşa water network, risk from DCAA exceeds the USEPA risk level 15.2-fold in women and 13.4-fold in men. However, since it was also found that the level of free chlorine in the network does not meet the required level, it should be noted that there may be an increase in the risk level if there is adequate chlorination in the supply.

INTRODUCTION

Chlorine is the most commonly used disinfectant in drinking water treatment due to its low cost, ease of use, and effectiveness in preventing waterborne disease. Chlorine kills or deactivates a large variety of waterborne microorganisms. However, since the mid-1970s, public health concerns about the disinfection process have increased. Studies have shown that chlorine reacts not only with waterborne parasitic pathogenic microorganisms, but also with natural organic matter (NOM), bromide, iodide, and nitrite in water to form trihalomethanes (THMs), haloacetic acids (HAAs), halonitromethanes (HNMs) and other disinfection by-products (DBPs) (Avşar et al., 2014; Avşar et al., 2015; Özdemir, 2020).

Studies on the formation of DBPs post-chlorination have shown that more than 700 species are formed in the water. However, in addition to these, there are also undetectable halogenated organic structures and it is clear that the undetectable compounds make up a significant portion of the total (Avşar et al., 2014; Avşar et al., 2015; Özdemir, 2021).

Given such potentially carcinogenic effects of some DBPs, DBP formation is the most important problem of the drinking water sector. Toxicological studies also show that DBPs can have adverse effects on development and reproduction. The potential health risks of DBPs have put pressure on industrialized countries to introduce maximum contaminant levels (MCL), in other words, limits, for types of DBPs in drinking water (Uyak et al., 2014).

HAAs is one of the important DBP groups and the most prevalent HAAs are MCAA (monochloroacetic acid), TCAA (trichloroacetic acid), and DCAA (dichloroacetic acid). The sum of MCAA, MBAA (monobromoacetic acid), DCAA, TCAA, and DBAA (dibromochloroacetic acid) is expressed as HAA_5. HAA_5 plus BCAA (bromochloroacetic acid), TBAA (tribromoacetic acid), BDCAA (bromodichloroacetic acid) and CDBAA (dibromochloroacetic acid) together makes up HAA_9. The United States Environmental Protection Agency's (USEPA) integrated risk information system has classified DCAA as a Group B2 (probable human carcinogen) and TCAA as a Group C (possible human carcinogen) component, and both components have been shown to be carcinogenic in animals (Avşar et al., 2014; Avşar et al., 2015; Thompson et al., 2015).

In 1998, because of the possible health risks, the maximum contaminant level of HAA_5 was set to $60~\mu g \cdot L^{-1}$ by the USEPA. In Canada, the limit has been set at $80~\mu g \cdot L^{-1}$ since 2008. The World Health Organization (WHO) has set up guidelines for chlorinated HAAs (MCAA: $20~\mu g \cdot L^{-1}$, DCAA: $50~\mu g \cdot L^{-1}$, TCAA: $200~\mu g \cdot L^{-1}$), but not for brominated HAAs. Neither the European Union nor Türkiye has set a limit value yet. However, the amount of HAA_9 in bromine-rich waters is 20–50% higher

WATER SA

than HAA$_5$, and it is important to consider HAA$_9$ in such waters because brominated species are potentially more carcinogenic than chlorinated species (Health Canada, 2008; Thompson et al., 2015).

Monitoring HAAs in the drinking water network is a complex process. Various parameters, such as the disinfection plan and chemicals used (chlorine or chlorine/chloramine or ozone/chlorine), water treatment processes, water source characteristics, bromide content, pH of chlorinated and distributed water, temperature, concentration of total and free chlorine, residence time, distribution system characteristics, and total organic carbon (TOC), affect the formation of HAAs. The formation kinetics, formation and stability of HAAs are still not completely understood. It is known that HAAs form faster than THMs, that some species decompose as pH increases, and that some species biodegrade in the absence of residual chlorine (Uyak et al., 2014; Thompson et al., 2015).

The formation of HAAs in chlorinated water varies considerably depending on the season and the length of the distribution system. These variations depend on the quality of the raw and treated water and on the operating parameters of the treatment plant as well. There are studies showing that the concentration of HAAs tends to first increase and then decrease with extended residence time in the water network (Uyak et al., 2014, Thompson et al., 2015). Because of all these reasons, monitoring seasonal changes in HAAs in drinking water systems as a function of the length of the distribution system is important for protecting public health and providing healthy drinking water.

In a study conducted by Uyak et al. (2014), seasonal and spatial variation in HAAs was observed in drinking water distribution networks fed by 3 different drinking water treatment plants in Istanbul. The highest median HAA$_9$ concentrations were observed in the spring and summer seasons, while the lowest concentrations were obtained in the autumn and winter periods. DCAA and TCAA were identified as the most significant HAA$_9$ components. It was observed that the seasonal average values of HAA$_9$ in all three distribution systems remained below 60 µg·L^{-1}, while the HAA$_9$ concentration in only one sample was measured to be 100.16 µg·L^{-1} during the sampling period.

According to a study that was conducted in Istanbul surface water sources (Büyükçekmece and Ömerli Lakes, between February 2010 and February 2011) by Avşar et al. (2014); the most important HAA$_9$ formation was caused by hydrophobic (HPO) and hydrophilic (HPI) fractions of NOM, respectively. According to Avşar et al. (2015), which was conducted for the same water sources as Avşar et al. (2014), organic substances with a molecular size below 1 kDa were found to be effective in the formation of HAA$_9$.

Uyak et al. (2007) studied Terkos, Büyükçekmece and Ömerli Lakes, Istanbul, with different bromide and organic matter concentrations, and observed the effect of this difference on DBP formation in terms of contact time, pH, chlorine dose and specific ultraviolet absorbance parameters. The predominant species of HAA was TCAA, followed by DCAA, with 60%, 49%, and 66% of total HAA in Terkos, Büyükçekmece, and Ömerli, respectively.

In the study of Ateş et al. (2007), a total of 29 surface waters from different regions of Türkiye were sampled seasonally and DBP formation potential was determined. The results showed that dissolved organic carbon (DOC) content was low in all water samples and ranged from 0.91 to 4.42 mg·L^{-1}. The ranges of annual average total THM and HAA concentrations were 21–189 and 18–149 µg·L^{-1}, respectively. Although the amount and distribution of DBPs vary by water source and season, chloroform and trichloroacetic acid (TCAA) are the most important species. The lack of a consistent trend in seasonal variations in water bodies suggests that the characteristics of dissolved organic matter (DOM) and its reactivity with chlorine vary seasonally in almost all water bodies.

Chen et al. (2008) investigated the formation potential for THMs and HAAs in a traditional water treatment plant in northern China. The highest concentrations of THMs and HAAs (each almost 500 µg·L^{-1}) were detected in autumn and the lowest in spring (no more than 100 µg·L^{-1}). Organic matter and algae were found to be significant DBP precursors. The hydrophobic acid fraction has the highest THM and HAA formation potential. Algae contribute about 20% to 50% of the total DBP formation potential during an algal bloom.

Gan et al. (2013) measured DBPs from the distribution systems of 10 water treatment plants in 3 provinces in China. The median of THM and HAA levels ranged from 0.7–62.7 µg·L^{-1} and 0.3–81.3 µg·L^{-1}, respectively. Chloroform, DCAA and TCAA were the dominant species observed in Guangzhou and Foshan water, while brominated THMs predominated in water from Zhuhai. THMs and HAAs showed clear seasonal variations with the total concentrations higher in winter than in summer.

Ghoochani et al. (2013) examined the variation of organic matter and HAA formation through samples collected from 3 major rivers (raw water) and 3 water treatment plant outlets (chlorinated water) supplied by these rivers in Tehran, Iran. Changes in the total organic matter (TOC), ultraviolet absorbance (UV$_{254}$), and specific ultraviolet absorbance (SUVA) were measured in raw water samples. HAAs were also measured in chlorinated water samples. The seasonal average concentration of TOC for the 3 rivers and HAAs for the 3 water treatment plants in were 4.00, 2.41 and 4.03 mg·L^{-1} and 48.75, 43.79 and 51.07 µg·L^{-1} for spring, summer and autumn, respectively. Seasonal variation indicated that HAA levels were much higher in spring and autumn, whereas TOC concentrations were similarly higher.

Golea et al. (2017) investigated the effect of DOM in THM and HAA formation in raw and treated surface waters. DBP formation potential in surface water sources has been studied with reference to the key water quality determinants (WQDs) of UV absorption (UV$_{254}$), colour, and DOM concentration. The DOM in the raw waters was found to comprise 30–84% (average 66%) of the more reactive HPO fraction, with this proportion falling to 18–63% (average 50%) in the treated water. Results suggest UV$_{254}$ to be as good an indicator of DBPFP as DOC or HPO for the raw waters, with values ranging from 0.79 to 0.82 for THMs and from 0.71 to 0.73 for HAAs for these three determinants. For treated waters the corresponding values were significantly lower, reflecting the lower HPO concentration.

Guilherme et al. (2014) investigated the occurrence of regulated and non-regulated DBPs in the drinking water of small systems in 2 provinces in Canada. It was reported that average measured concentrations of these compounds was much higher than that reported in the literature for medium and large systems. The measured average value for THMs was 75 µg·L^{-1} and for HAAs was 77 µg·L^{-1}. Investigated DBPs decreased between the autumn and winter and then increased to eventually reach a maximum in late summer.

Rodriguez et al. (2004) investigated the seasonal and spatial variation of post-chlorination DBPs in a drinking water distribution system located in a region where very significant seasonal variations in water temperature and surface water quality occur. The analysis of a large number of collected samples showed that the seasonal and geographical variations of THMs and HAAs were of particular importance in this region. THM levels were found to be about 5 times higher on average in summer and autumn than in winter, while average HAA levels were about 4 times higher in spring than in winter.

Rodriguez et al. (2007) studied HAA formation during the treatment process in 2 facilities where surface water is pre-chlorinated. Samples were collected over 1 year at 4 stations in order to investigate both seasonal and spatial variations of HAAs. The results revealed that the formation of HAAs was highest during the pre-chlorination process, where precursor compounds and pre-chlorination dose were both higher and more variable, and that formation lasted from the pre-chlorination point until the settled water occurred, due to remaining levels of residual chlorine. It was identified that DCAA was the predominant HAA species.

To understand water quality variability in small systems, Scheili et al. (2015) monitored water quality in 25 small municipal systems in 2 Canadian regions from the water source to the end of the distribution system. The database obtained was used to create a global portrait of physical, chemical, and microbiological water quality parameters. It was observed that the values of DOM varied during the different seasons, with maxima in autumn for both provinces. In the regions studied, the highest THM and HAA values were reached in the warmer seasons (summer, autumn), as observed in previous studies with large systems. Observed THM and HAA levels were 3 times higher in systems in the province of Newfoundland and Labrador than in the province of Quebec.

Serodes et al. (2003) chlorinated seasonally treated waters from 3 major drinking water supplies in the Quebec City region and studied the formation of THM and HAA. They concluded that chloroform was the predominant compound for THM species, while dichloroacetic acid and trichloroacetic acid were the most abundant for DBP species due to low bromide concentrations in the raw waters. Significant differences in DBP formation were observed, mainly due to the type of disinfectant used for raw water in the three utilities. The use of pre-ozonation as opposed to pre-chlorination (or direct chlorination) in one of the utilities appears to be the primary factor contributing to that utility's ability to meet current THM and future HAA standards.

In this study, HAA formation and health risk via ingestion were investigated in 2 networks fed by groundwater and surface water, where the organic matter level is low, when chlorination is not properly utilized (under uncontrolled conditions). HAAs are of special importance in these networks, since the networks are short and HAAs are formed faster than other DBPs. In addition, no

study has been found in the literature evaluating the formation of HAA under insufficient chlorination conditions in groundwater with low NOM levels. There is also no study apparent in the literature on whether HAA formation will pose a risk in low organic content waters under insufficient chlorination conditions. In this study, answers to all these questions were sought. Although the formation of HAA is important and may pose a risk in Türkiye, there is no limit value being applied yet.

MATERIALS AND METHODS

Sampling points

This study investigated the quality of raw water sources used in Muş Province city centre, the seasonal and spatial variance of HAAs formed after chlorination of these sources, and the risk to human health via ingestion. Therefore, sampling and analytical studies were conducted on the dates of 20 July 2017 (summer); 26 October 2017 (autumn); 8 January 2018 (winter); and 7 March 2018 (spring) in the city centre of Muş Province, to characterize the summer, autumn, winter, and spring seasons. Figure 1 shows the location of Muş, networks and sampling points used in the study.

The drinking water network for Muş city centre is provided from groundwater extracted by 17 existing boreholes around the main pumping station (Point 1 in Fig. 1). The water taken through the boreholes is pumped to the tank of the main pumping station, and released to the water network after chlorination via liquid sodium hypochlorite solution (NaOCl). There is also another small network in the city centre. Water from a mountain spring (surface water) is fed to the Muratpaşa water tank, chlorinated with NaOCl, and pumped to the small water network (Point 5 in Fig. 1). Figure 2 shows photographs of the water tanks.

The characteristics of water networks and sampling points are shown in Table 1. The points in Table 1 are arranged in the order relating to the water path between source and water network.

It was observed that the water drawn from 17 boreholes in the vicinity of the main pumping station, Station 1, reached the main pumping station where it was mixed in the tank, chlorinated, and sent to the Reservoir 3 at Point 9 and released to the network. Seasonal sampling and analyses were conducted at Points 2, 9, 7, 10, 4 and 3 of this network. Additionally, seasonal sampling
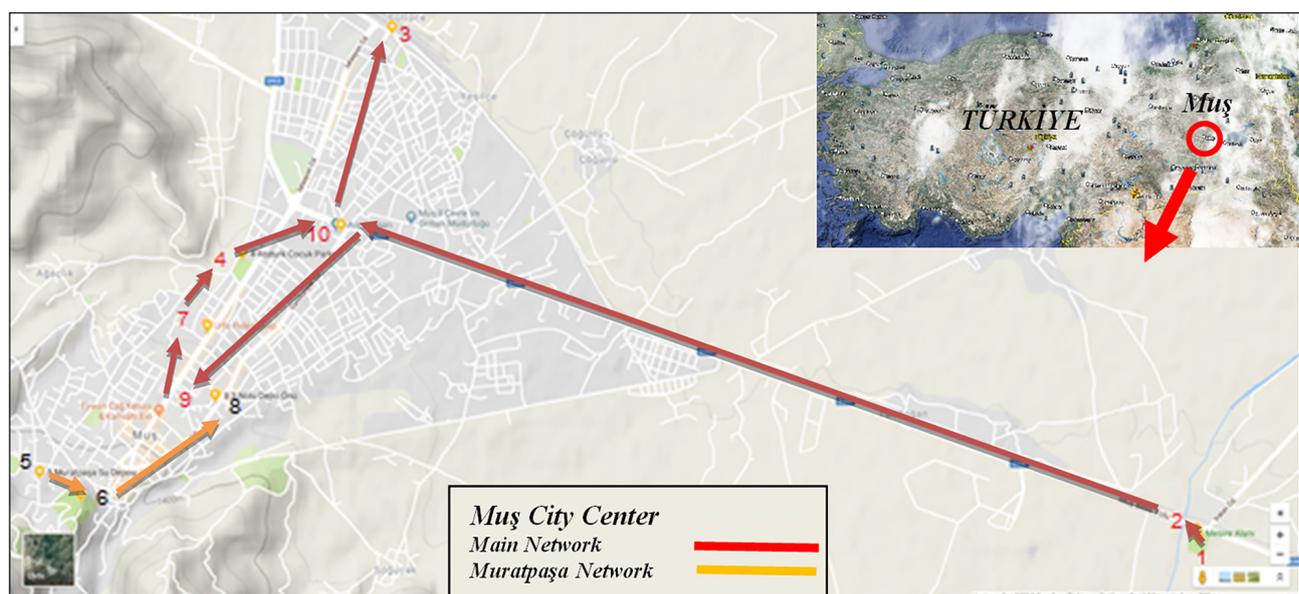
Figure 1. Networks and sampling points in the center of Muş Province

Figure 2. Photographs showing the reservoirs where the study was conducted: (a) external view of main pumping station; (b) chlorine dosage pump inside main tank; (c) main water tank inside; (d) Muratpaşa water tank

Table 1. Description of sampling points

No.	Name of point	Type of sample	Coordinates
	Main reservoir and water network		
1	Main reservoir	Raw water (chlorinated)	38.727294; 41.580537
2	Main reservoir outlet	Chlorinated tap water	38.727801; 41.580370
9	Muş centre, inner side of reservoir 3	Chlorinated tap water	38.736459; 41.496992
7	Central restaurant	Chlorinated tap water	38.740921; 41.496384
4	Central Atatürk playground	Chlorinated tap water	38.745588; 41.499400
10	Central bus terminal garden	Chlorinated tap water	38.747446; 41.507699
3	Final point of water network	Chlorinated tap water	38.760184; 41.512072
	Muratpaşa reservoir and its water network		
5	Muratpaşa reservoir outlet	Raw water (chlorinated)	38.731482; 41.482136
6	Central Kale park	Chlorinated tap water	38.730002; 41.485636
8	Front of central reservoir 3	Chlorinated tap water	38.736480; 41.497141

and analyses were conducted for Muratpaşa reservoir, where the mountain spring was located, and at Points 6 and 8 of the network supplied from this reservoir. HAAs from post-chlorination DBPs in the reservoirs and water distribution systems of 2 different distribution networks were monitored. The sampling points in both networks are listed in Table 1, give in order from the source to the final distribution point.

One-time sampling was conducted in the context of 4 different seasons from 2 networks consisting of 10 points in total. Water was wasted for at least 1 min in each fountain before sampling. Samples were taken in polyethylene sample containers and transferred to the laboratory for further analysis.

Analytical methods

As part of the study, the water quality parameters and post-chlorination–formed HAA species were measured in 4 seasons in

2 water reservoirs and water networks where the existing water is supplied in the reservoirs in the city centre of Muş.

The parameters measured, the devices used and the measurement methods of the devices are given in Table 2. Detailed analytical methods are given in Avşar and Toröz (2018); Avşar et al. (2014); Avşar et al. (2015) and Avşar et al. (2020).

Conductivity, pH, ORP, free and total chlorine, and turbidity were measured on-site after sampling. Than samples were dechlorinated with 0.1 mL of 100 mg·mL^{-1} Na$_2$S solution and transferred to the laboratory for further analysis, given in Table 2.

HAA analysis was conducted according to USEPA 552.3 liquid-liquid micro-extraction method via a GC equipped with μECD detector. The method consists of sample extraction and methyl ester conversion of HAA species. Further information about device, method and detection limits is given in Avşar and Toröz (2018).

Table 2. Parameters measured, devices and measurement methods utilized

Parameter	Device	Method	References
TOC (total organic carbon)	Teledyne Tekmar Torch TOC/TN Analyzer	Standard Methods 5310-B	Avşar and Toröz, 2018 Avşar et al., 2014 Avşar et al., 2015 Avşar et al., 2020
TN (total nitrogen)		Standard Methods 4500N-B	
pH/temperature	Hach Hq40d Multimeter	USEPA Electrode Method 8156	
Conductivity		USEPA Direct Measurement Method 8160	
ORP (oxidation reduction potential)		Direct Measurement Method 10228	
HAAs	Agilent 6890 GC-µECD	USEPA Method 552.3	
Free chlorine	Lovibond Comparator	Standard Methods 4500 Cl-G	
Total chlorine			
Nitrate	WTW Photolab 7600 UV-VIS Spectrophotometer	Standard Methods 4500 NO_3^--B	
Bromide		Standard Methods 4500 Br-B	
Alkalinity	Titrimetric	Standard Methods 2320-B	
Turbidity	WTW Turb 355 IR	ISO 7027 – DIN/EN 27 027	
UV_{254}	WTW Photolab 7600 UV-VIS Spectrophotometer	Standard Methods 5910-B	

RESULTS

The research carried out consisted of 2 stages. In the first stage, sampling and analysis were carried out for 2 different networks. In the second stage, according to the HAA analysis results obtained in the first stage, risk analysis was conducted to determine whether drinking these waters poses a human health risk.

Analytical results

The water quality parameters for the samples taken from the main reservoir and its network in the 4 seasons are compared with Turkish Standards (TS 266, 2005: water intended for human consumption) in Table 3. Minimum, maximum and mean values for the main reservoir are given in Table 4.

A comparison of seasonal results for Muratpaşa water reservoir and its network with Turkish Standards of is given in Table 5. Minimum, maximum and mean values of the results for Muratpaşa network are given in Table 6.

HAA concentrations recorded in 2 reservoirs and their networks are given in Tables 7 and 8.

Sampling points for both water supplies are listed in the tables in an order that matches the route the water takes in the water network.

When the water quality results are examined, it can be seen that chlorination was insufficient in both water sources. Free chlorine was detected at the main reservoir outlet (Point 2) in 4 seasons (min: 0.2; max: 1.3 and mean: 0.9 mg·L⁻¹). However, in general, free chlorine in the main network is depleted as it goes from reservoir outlet (Point 2) to network end-point (Point 3). At Point 3, no free chlorine was detected in the water during any sampling period (Tables 3–4).

Except for the last point (Point 8: min: 0; max: 0.6; mean: 0.2 mg·L⁻¹), no chlorine was found in the network during the sampling period in the Muratpaşa reservoir and network (Tables 5–6). This is related to frequent power cuts in the city. When the electricity is off, the chlorine dosing system is disabled. The dosing system is not activated automatically when the electricity is re-supplied to the system. Manual start-up of the dosing system is required. This prevents sufficient chlorine dosing to the system.

TOC (min: 0.0021; max: 0.3553; mean: 0.1787 mg·L⁻¹), TN (min: 1.9166; max: 2.4552; mean: 2.2773 mg·L⁻¹) and turbidity (min: 0.10; max: 0.65; mean: 0.27 NTU) values in the main reservoir were generally at low levels. It was observed that the nitrate value (min: 2.00; max: 4.80; mean: 3.55 mg·L⁻¹) remained below the TS 266 limit value throughout the monitoring period. The UV_{254} parameter (min: 0.002; max: 0.0140; mean: 0.0063 cm⁻¹), which is an indicator of the organic content in the water, is similarly low, along with the TOC values. These results indicate that there is no significant pollution of the groundwater, in general.

TOC (min: 0.1088; max: 0.3052; mean: 0.2070 mg·L⁻¹) and TN values (min: 0.6358; max: 1.0441; mean: 0.8729 mg·L⁻¹) in the Muratpaşa reservoir were also low. However, from time to time, the limit value is exceeded in terms of turbidity in the reservoir (max: 4.19 NTU) and network (max: 2.11 NTU). It is thought that this situation is due to the leakages that occur due to the age of the pipelines.

The low UV_{254} values in both networks indicate low organic content in the waters. There was no significant change in conductivity or ORP values either. There was no significant change in water quality parameters at the measurement points of both networks. It is thought that the changes at specific measurement points that can be seen in the parameters from time to time are related to the network leakages that occur from time to time at these points.

USEPA National Primary Drinking Water Regulation (1994) states that in order to prevent DBP formation, the TOC concentration in the water should be reduced to below 2 mg·L⁻¹ before chlorination. However, although the TOC and UV_{254} parameters in both networks are considerably lower than the values specified in the publications referenced in the introduction, it was determined that the USEPA HAA_5 limit value of 60 µg·L⁻¹ is exceeded (max: 68.87 µg·L⁻¹) in the main network at times. HAA_9 values reach values of up to 91.54 µg·L⁻¹. In Muratpaşa network, the HAA_5 limit value was not exceeded. However, despite the lack of sufficient chlorine in the water, concentrations close to the limit value (max HAA_5: 54.95 µg·L⁻¹) were determined, while maximum HAA_9 concentration was determined as 55.07 µg·L⁻¹. On the other hand, MCAA can occasionally exceed the WHO limit value of 20 µg·L⁻¹ in both networks. This shows that there is potential for HAA formation in the case of adequate chlorination of the water, and that the water includes precursor compounds for DBP formation.

Table 3. All-season water quality parameters for samples taken at the points determined in the main pumping station and the connected water network

Parameter		pH	Conductivity	ORP	Temp.	Free chlorine	Total chlorine	Alkalinity	Nitrate	Bromide	Turbidity	UV$_{254}$	TOC	TN
Unit		-	µS·cm^{-1}	mV	°C	mg·L^{-1}	mg·L^{-1}	mgCaCO$_3$·L^{-1}	mg·L^{-1}	mg·L^{-1}	NTU	cm^{-1}	mg·L^{-1}	mg·L^{-1}
Point 1	1	6.98	285	222	15.8	0	0	135.1	2	<0.1	0.11	0.014	0.3553	1.9166
	2	7.58	313	215.7	12.2	0	0	116.04	3.2	0.16	0.10	0.002	<0.05	2.4552
	3	7.87	412	415	8.5	0	0	154	4.2	0.37	0.21	0.005	<0.05	2.4267
	4	7.75	473	174.2	11.4	0	0	154	4.8	0.44	0.65	0.004	0.002	2.311
Point 2	1	7.08	362	561.5	16.1	0.8	0.8				0.56	0.001	0.3791	2.9867
	2	7.4	323	325.9	14.3	1.1	1.1				0.45	0.002	<0.05	3.2475
	3	7.71	395	528.5	10.2	0.2	0.2				0.02	0.003	0.1824	2.5507
	4	7.79	473	670.9	12.2	1.3	1.5				0.83	0.001	<0.05	2.2882
Point 9	1	7.41	357	620	20.01	0.9	1.1				0.24	0.027	0.381	2.7268
	2	7.16	425	213.1	14.9	0	0				0.63	0.009	<0.05	2.8859
	3	7.88	369	184.3	8.4	0	0				0.11	0.001	<0.05	2.6631
	4	7.64	346	115.5	12.1	0	0				0.86	0.003	<0.05	2.4572
Point 7	1	7.39	356	529.5	17.7	0.2	0.2				0.41	0.004	0.0953	2.6179
	2	7.44	382	151.2	14.2	0	0				0.57	0.002	<0.05	2.2629
	3	7.82	404	163.7	11.3	0	0				0.11	0	<0.05	1.999
	4	7.58	422	231.4	13	0	0				4.34	0.001	<0.05	1.5233
Point 4	1	7.05	682	230.5	20.1	0	0				0.18	0.002	0.3975	5.6338
	2	7.46	375	135	14.8	0	0				1.43	0.001	<0.05	2.5835
	3	7.9	413	211.6	9.9	0	0				0.11	0	0.3376	2.2249
	4	7.38	477	180.5	12	0	0				3.86	0.003	0.9574	1.9761
Point 10	1	7.28	353	574.9	17.2	0.5	0.7				0.41	0.009	0.3421	2.8497
	2	7.34	374	140.6	15.4	0.1	0.1				0.59	0.003	0.2221	3.0244
	3	8.23	416	214.2	9.4	0	0				0.27	0.005	0.9126	2.6973
	4	7.78	361	194.2	12.3	0	0				1.03	0.009	<0.05	2.4904
Point 3	1	7.24	375	368.1	24.9	0	0				0.32	0.007	0.3244	2.9443
	2	7.23	754	162.2	17.8	0	0				0.29	0.002	<0.05	7.4393
	3	7.87	397	197.4	8.3	0	0				0.04	0.003	0.1005	2.8998
	4	8.38	475	178.3	12.2	0	0				0.65	0.001	<0.05	2.3424
TS 266 Limits	-2005	6.5–9.5	-	-	-	Is desirable to be at least 0.2–0.5 mg·L^{-1}				50	1	-	-	-

1 – summer, 20 July 2017; 2 – autumn, 26 October 2017; 3 – winter, 8 January 2018; 4 – spring, 7 March 2018

Table 4. The minimum, maximum and mean values of the all-season water quality parameters of the samples taken at the points determined in the main pumping station and the connected water network

Parameter		pH	Conductivity	ORP	Temp.	Free chlorine	Total chlorine	Alkalinity	Nitrate	Bromide	Turbidity	UV$_{254}$	TOC	TN
Unit		-	µS·cm^{-1}	mV	°C	mg·L^{-1}	mg·L^{-1}	mgCaCO$_3$·L^{-1}	mg·L^{-1}	mg·L^{-1}	NTU	cm^{-1}	mg·L^{-1}	mg·L^{-1}
Point 1	min	6.98	285	174.2	8.5	0.0	0.0	116.0	2.00	0.16	0.10	0.002	0.0021	1.9166
	max	7.87	473	415.0	15.8	0.0	0.0	154.0	4.80	0.44	0.65	0.0140	0.3553	2.4552
	mean	7.55	371	256.7	12.0	0.0	0.0	139.8	3.55	0.32	0.27	0.0063	0.1787	2.2773
Point 2	min	7.08	323	325.9	10.2	0.2	0.2				0.02	0.0010	0.0000	2.2882
	max	7.79	473	670.9	16.1	1.3	1.5				0.83	0.0030	0.3791	3.2475
	mean	7.50	388	521.7	13.2	0.9	0.9				0.46	0.0018	0.1872	2.7683
Point 9	min	7.16	346	115.5	8.4	0.0	0.0				0.11	0.0010	0.0000	2.4572
	max	7.88	425	620.0	20.0	0.9	1.1				0.86	0.0270	0.3810	2.8859
	mean	7.52	374	283.2	13.9	0.2	0.3				0.46	0.0100	0.1905	2.6833
Point 7	min	7.39	356	151.2	11.3	0.0	0.0				0.11	0.0000	0.0000	1.5233
	max	7.82	422	529.5	17.7	0.2	0.2				4.34	0.0040	0.0953	2.6179
	mean	7.56	391	269.0	14.1	0.1	0.1				1.36	0.0018	0.0477	2.1008
Point 4	min	7.05	375	135.0	9.9	0.0	0.0				0.11	0.0000	0.3376	1.9761
	max	7.90	682	230.5	20.1	0.0	0.0				3.86	0.0030	0.9574	5.6338
	mean	7.45	487	189.4	14.2	0.0	0.0				1.39	0.0015	0.5642	3.1046
Point 10	min	7.28	353	140.6	9.4	0.0	0.0				0.27	0.0030	0.0000	2.4904
	max	8.23	416	574.9	17.2	0.5	0.7				1.03	0.0090	0.9126	3.0244
	mean	7.66	376	281.0	13.6	0.2	0.2				0.58	0.0065	0.3692	2.7655
Point 3	min	7.23	375	162.2	8.3	0.0	0.0				0.04	0.0010	0.0000	2.3424
	max	8.38	754	368.1	24.9	0.0	0.0				0.65	0.0070	0.3244	7.4393
	mean	7.68	500	226.5	15.8	0.0	0.0				0.32	0.0033	0.1416	3.9065

Table 5. All-season water quality parameters analysis results of the samples taken at the points determined in Muratpaşa reservoir and the connected water network

Parameter		pH	Conductivity	ORP	Temp.	Free chlorine	Total chlorine	Alkalinity	Nitrate	Bromide	Turbidity	UV$_{254}$	TOC	TN
Unit		-	µS·cm^{-1}	mV	°C	mg·L^{-1}	mg·L^{-1}	mgCaCO$_3$·L^{-1}	mg·L^{-1}	mg·L^{-1}	NTU	cm^{-1}	mg·L^{-1}	mg·L^{-1}
Point 5	1	7.39	323	200.5	21.1	0	0	177.6	1.1	0.12	0.23	0.005	0.3052	0.8554
	2	6.82	335	183.6	11.1	0	0	181.796	1.5	0.15	0.28	0.004	<0.05	0.6358
	3	7.54	455	249	3.6	0	0	230	2.2	0.4	0.09	0	<0.05	0.9564
	4	7.54	433	141.2	10.1	0	0	154	3.4	0.44	**4.19**	0.002	0.1088	1.0441
Point 6	1	7.56	360	180.6	22.7	0	0				0.35	0.014	0.3384	0.8704
	2	7.02	327	212.7	14.5	0	0				0.24	0.006	<0.05	0.6658
	3	6.26	402	256.3	5	0	0				0.01	0	<0.05	0.8775
	4	7.36	353	117.1	8	0	0				**2.11**	0.006	0.2549	0.9992
Point 8	1	7.43	352	596	16.9	**0.6**	**0.8**				0.22	0.006	0.4522	2.4642
	2	7.13	360	219.8	13.3	0	0				0.75	0.006	<0.05	3.0502
	3	8.26	394	158.1	7.2	0	0				0	0.001	<0.05	2.6618
	4	7.68	424	115	12	0	0				0.84	0	0	2.3894
TS 266 limits	-2005	6.5–9.5	-	-	-	Is desirable to be at least 0.2–0.5 mg·L^{-1}			50	-	1	-	-	-

1 – summer, 20 July 2017; 2 – autumn, 26 October 2017; 3 – winter, 8 January 2018; 4 – spring, 7 March 2018

Table 6. The minimum, maximum and mean values of the all-season water quality parameters of the samples taken at the points determined in the Muratpaşa Water reservoir and the connected water network

Parameter		pH	Conductivity	ORP	Temperature	Free chlorine	Total chlorine	Alkalinity	Nitrate	Bromide	Turbidity	UV$_{254}$	TOC	TN
Unit		-	µS·cm^{-1}	mV	°C	mg·L^{-1}	mg·L^{-1}	mgCaCO$_3$·L^{-1}	mg·L^{-1}	mg·L^{-1}	NTU	cm^{-1}	mg·L^{-1}	mg·L^{-1}
Point 5	min	6.82	323.00	141.2	3.6	0.0	0.0	154.0	1.1	0.12	0.09	0.000	0.1088	0.6358
	max	7.54	455.0	249.0	21.1	0.0	0.0	230.0	3.4	0.44	4.19	0.005	0.3052	1.0441
	mean	7.32	386.50	193.6	11.5	0.0	0.0	185.8	2.1	0.28	1.20	0.003	0.2070	0.8729
Point 6	min	6.26	327.00	117.1	5.0	0.0	0.0				0.01	0.000	0.2549	0.6658
	max	7.56	402.00	256.3	22.7	0.0	0.0				2.11	0.014	0.3384	0.9992
	mean	7.05	360.50	191.7	12.6	0.0	0.0				0.68	0.007	0.2967	0.8532
Point 8	min	7.13	352.00	115.0	7.2	0.0	0.0				0.00	0.000	<0.05	2.3894
	max	8.26	424.00	596.0	16.9	0.6	0.8				0.84	0.006	0.4522	3.0502
	mean	7.63	382.50	272.2	12.4	0.2	0.2				0.45	0.003	0.2261	2.6414

Table 7. All-season HAA analyses of samples taken from the points determined in the main pumping station and connected water network

Parameter		MCAA	MBAA	DCAA	BCAA	TCAA	DBAA	BDCAA	CDBAA	TBAA	HAA$_5$	HAA$_9$
Unit							µg·L^{-1}					
Point 1	1	*	0.59	1.89							2.48	2.48
	2		0.49	1.91							2.40	2.40
	3											
	4											
Point 2	1	**39.51**	0.75				0.40				40.66	40.66
	2	**59.68**	0.86	3.43	2.34					25.23	**63.97**	91.54
	3	1.86									1.86	1.86
	4											
Point 9	1	**44.39**	0.66	12.49	0.44		0.20				57.74	58.18
	2		0.50	2.38							2.89	2.89
	3											
	4											
Point 7	1	**41.76**	0.65	2.77	0.60		0.25				45.43	46.03
	2		0.06	1.97							2.04	2.04
	3											
	4	0										
Point 4	1	**64.27**	1.34	3.27						13.28	**68.87**	82.15
	2		0.06	2.24							2.30	2.30
	3	2.40					1.38				3.79	3.79
	4											
Point 10	1		0.24	1.96							2.20	2.20
	2	**37.73**	0.51	0.59			0.40				39.22	39.22
	3											
	4											
Point 3	1	**40.71**	0.74	2.75			0.37				44.56	44.56
	2		0.22	2.27							2.49	2.49
	3											
	4											

Empty cells mean compound or group not detectable (ND)
1 – summer, 20 July 2017; 2 – autumn, 26 October 2017; 3 – winter, 8 January 2018; 4 – spring, 7 March 2018

Table 8. All-season HAA analyses of samples taken from the points determined in the Muratpaşa reservoir and connected water network

Parameter		MCAA	MBAA	DCAA	BCAA	TCAA	DBAA	BDCAA	CDBAA	TBAA	HAA5	HAA9
Unit						μg·L⁻¹						
Point 5	1	**36.28**	1.79	2.58	*						40.66	40.66
	2		0.05	2.08							2.13	2.13
	3											
	4											
Point 6	1	**41.87**	1.77	0.00		0.18					43.83	43.83
	2		0.21	2.29							2.50	2.50
	3											
	4											
Point 8	1	**43.66**	0.65	10.17	0.12	0.47					54.95	55.07
	2		0.26	2.21							2.47	2.47
	3											
	4											

Empty cells mean compound or group not detectable (ND)
1 – summer, 20 July 2017; 2 – autumn, 26 October 2017; 3 – winter, 8 January 2018; 4 – spring, 7 March 2018

The fact that there is not enough chlorine in the water most of the time made it impossible to make a seasonal assessment of HAA formation. In both networks, HAA components could not be detected in the water in the winter and spring periods. Likewise, although the amount of bromide in both water supplies is highest in the winter and spring periods, the absence of HAA detection during these periods makes it impossible to evaluate the effect of bromide on HAA formation potential.

In both networks, the most important HAA component was determined as MCAA. In general, the irregularity of chlorination in the networks prevents the evaluation of the residence time in the both networks. However, considering the summer period in which HAA types were detected in both networks, the highest HAA5 and HAA9 concentrations were determined towards the end in the main network and at the last point in the Muratpaşa network.

The low bromide level in both water sources limits the formation of brominated HAAs in the water. Brominated species were not detected in water for most of the sampling period.

Risk assessment

Despite the lack of sufficient chlorination and the low organic matter content in both networks, it was observed that HAA species could form and exceed the limit values from time to time. In this context, the effect of consuming these waters on public health in terms of HAA compounds was determined through a risk assessment.

The measurement of cancer risk for DBPs is performed from 3 perspectives: through oral ingestion, dermal absorption, and inhalation exposure. However, since there are only risk factors for oral ingestion of HAA components (DCAA and TCAA) in the literature, it is not possible to perform measurements for dermal absorption and inhalation exposure. The oral ingestion-related risk is calculated using the following equation (Gan et al., 2013; Avşar et al., 2020):

$$CR = CDI_{oral} \times PF_{oral} \qquad (1)$$

$$CDI_{oral} = \frac{(CW \times IR \times EF \times ET)}{(BW \times AT)} \qquad (2)$$

where: C_R is cancer risk from HAA component ingestion; CDI_{oral} is chronic daily ingestion of that species (oral: mg·kg⁻¹·day⁻¹); PF_{oral} is potential factor or slope factor (oral: mg·kg⁻¹·day⁻¹)

Among the HAAs examined in the study, there are PF_{oral} values for DCAA and TCAA reported in the literature as 0.05 and 0.07,

respectively (Gan et al., 2013). The variables in Eq. 2 and the values used to calculate them are listed in Table 9.

Given the variables in Table 9, the measured risk values for the highest concentrations obtained in 2 different water supplies for DCAA and TCAA are given in Table 10.

According to the EPA, a probability of occurrence of an event of more than 1 in 1 million means that the minimum or negligible risk level for that event has been exceeded (Gan et al., 2013; Avşar et al. 2020). In this context if the estimated C_R is greater than 1 million it indicates a health risk. Alternatively, multiple rate (M_R) values can be estimated using Eq. 3.

$$M_R = \frac{C_R}{(1 \times 10^6)} \qquad (3)$$

The risk assessment considered the highest concentrations detected in the network to determine the worst case for the HAA compounds that were used as the basis for the assessment. In this context, the risk level related to DCAA in the main network was found to be 18.7 times higher for women and 16.5 times higher for men, based on the components for which a risk assessment was performed. Since TCAA is not detectable in the network, there is no risk in relation to this component.

In the Muratpaşa water network, on the other hand, it was observed that the maximum concentration determined for DCAA exceeds the risk level 15.2-fold for women and 13.4-fold for men. The concentration determined in relation to TCAA does not pose a risk for men or women. However, since it was found that the level of free chlorine in the network does not meet the required level, it should be noted that there may be an increase in the risk level if there is adequate chlorination in the supplies. The risk assessment, therefore, should be repeated using values obtained after adequate chlorination in the system.

Quantitative differences in the variables underlying the calculation for men and women lead to different results, so that the risk values calculated for women are higher at the same concentration.

CONCLUSIONS AND RECOMMENDATIONS

In the study, seasonal changes in water quality parameters in 2 water sources and connected water supplies in Muş city center, and HAA formation as a result of chlorination, were monitored. Both water supplies have undergone many revisions since their construction, but it was not possible to obtain a plan or project on which these revisions were plotted. The networks need to be

Table 9. Values based on risk assessment

Variable	Description	Unit	Accepted value in this study	Reference
CW	Concentration of chemical (HAA component) in water	mg·L^{-1}	The highest concentrations measured for the species in 2 different networks to calculate the maximum risk	Gan et al., 2013
IR	Ingestion rate	L·day^{-1}	2	Gan et al., 2013
EF	Exposure frequency	day·yr^{-1}	365	Gan et al., 2013
ET	Exposure time	yr	According to TSI statistics for the period of 2014–2016, life expectancy at birth in Turkey is generally 78 years, with an average of 75.3 years for men and 80.7 years for women.	TSI 2018
BW	Body weight	kg	According to TUIK 2010 data, the average weight in Türkiye was determined as 71.5 kg. While the average weight was 75.8 kg for men, it was determined as 66.9 kg for women.	TSI 2010
AT	Average time	day	Average life expectancy expressed in days	TSI 2018

Table 10. Risk assessment results

Network	Contaminant	DBP concentration (mg·L^{-1})	C_R for women	M_R for women	C_R for men	M_R for men
Main network	DCAA	12.48532 x 10^{-3}	1.8674 x 10^{-5}	18.7	1.647135 x 10^{-5}	16.5
	TCAA	-	-	-	-	-
Muratpaşa network	DCAA	10.17277 x 10^{-3}	1.52057 x 10^{-5}	15.2	1.342205 x 10^{-5}	13.4
	TCAA	0.18424 x 10^{-3}	3.85549 x 10^{-7}	0.4	3.40284 x 10^{-7}	0.3

observable and controllable for the distribution of healthy water. Therefore, a sound network plan should be created, first and foremost.

The first phase of the study consisted of water quality analyses. Chlorine, turbidity, pH, temperature, alkalinity, TOC, TN, ammonia, bromide, UV$_{254}$, conductivity and ORP parameters were measured.

It was found that several quality parameters can sometimes be outside the limit range given in TS 266. During the monitoring period, adequate and necessary chlorination was not performed in both water supplies. In this context, the chlorination process carried out by the Muş Municipality in the reservoirs and in the network should be monitored regularly. In addition, due to the frequent power outages, it would be beneficial to support the chlorination plant with a generator.

It was observed that the pH falls below 6.5 in winter at Point 6 of the Muratpaşa network. This is believed to be a local problem that does not affect the entire network, and it would be beneficial to periodically review the network to identify such local problems and develop solutions.

Turbidity levels exceeded the limit at some locations in both supplies and in the Muratpaşa reservoir, especially in the spring season. The high turbidity value in the Muratpaşa reservoir indicates that the residence time is not sufficient to allow for solids to settle out or that the reservoir is contaminated or not sufficiently cleaned.

In the second phase of the work, seasonal and spatial monitoring for HAAs in the network was conducted. However, since no chlorine was detected in the water most of the time during the study period, it was not possible to obtain sufficient data to determine the HAA formation potential in the water or the spatial variation. In this case, it will be useful to determine the HAA formation potential in the laboratory by taking raw water from both reservoirs on a seasonal basis.

Given the obtained HAA measurements, it was noted that:

- The HAA$_5$ concentration in the central network exceeds the limit from time to time, while the limit is not exceeded in Muratpaşa, but HAA$_5$ levels may exceed the limits if the necessary chlorination is carried out in both waters.
- The limits for MCAA given by the World Health Organization are exceeded from time to time.
- Currently, there appears to be a potential risk to women and men from both supplies with respect to the DCAA component via ingestion.

AUTHOR CONTRIBUTIONS

EA (Associate Professor) wrote the manuscript, conducted all the experiments and visited facilities. AK (MsD student) wrote the manuscript and conducted the experiments.

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