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Evaluation of formation and health risks of disinfection by-products in drinking water supply of Ggaba waterworks, Kampala, Uganda

Annitah Nshemereirwe[a,b,*], Feleke Zewge[c] and Enos Malambala[b]

[a] College of Natural and Computational Sciences, Addis Ababa University, Addis Ababa, Ethiopia
[b] National Water and Sewerage Corporation Ggaba Water Quality Laboratory, Kampala, Uganda
[c] Africa Center of Excellence for Water Management, Addis Ababa, Ethiopia
*Corresponding author. E-mail: anshemereirwe365@gmail.com

EM, 0000-0003-4187-4161

ABSTRACT

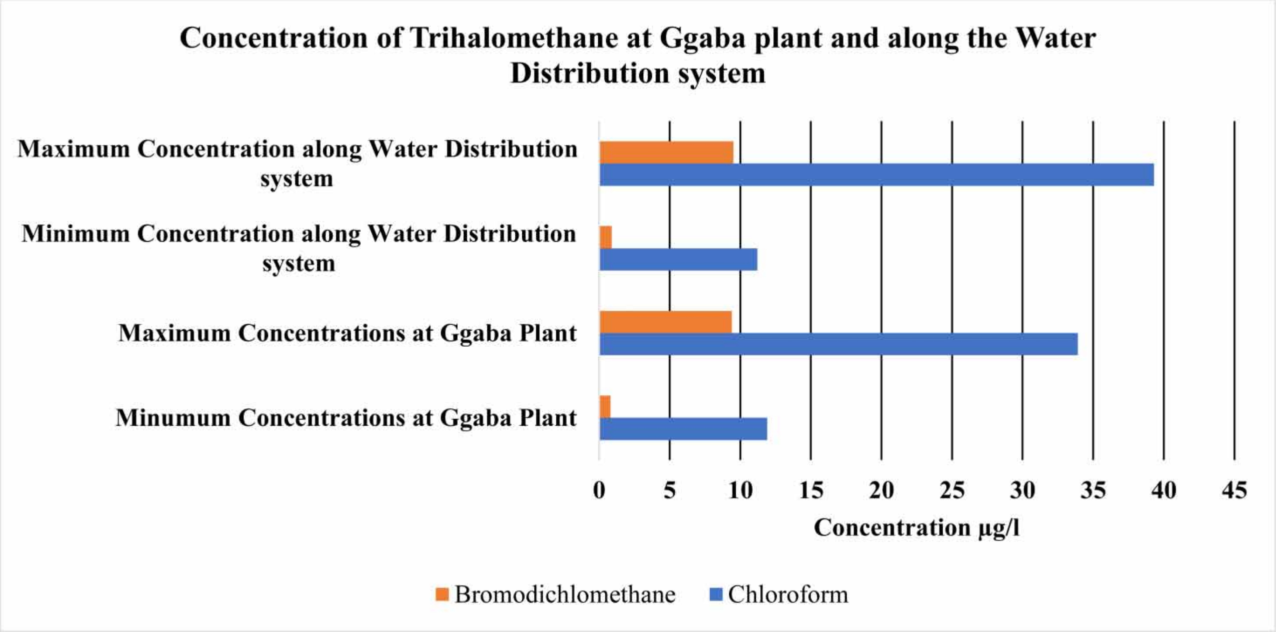
In developing countries, the evaluation of disinfection by-products in drinking water has been neglected because most water utility companies focus on microbial elimination. As a result, this study aimed at evaluating trihalomethane formation, the relation between water quality parameters and trihalomethane formation, and the estimation of the associated potential health risks in drinking water. The headspace purge and trap coupled with a gas chromatography–mass spectrometry technique was used to quantify trihalomethane. The concentrations of trihalomethane found in the water were within the National Water and Sewerage Corporation, World Health Organization guidelines, and the United States Environmental Protection Agency standards. Total organic carbon, ultraviolet absorbance at 254 nm, bromide concentration, and the temperature had a positive and significant correlation, pH had a positive but non-significant correlation, while the residual chlorine had a negative but significant correlation with trihalomethane formation. The potential health risk using the WHO index was 0.4, indicating no noncarcinogenic risk to human health in the study area. The lifetime carcinogenic risks of trihalomethane due to oral ingestion, dermal, and inhalation were 2.5×10^{-5}, 9.1×10^{-6}, and 8.3×10^{-6} for females and 2.4×10^{-5}, 1×10^{-5}, and 7.9×10^{-6} for males, and the values were within the USEPA acceptable low-risk range of $1\times10^{-6}<\mathrm{CR}<5.1\times10^{-5}$.

Key words: disinfection by-products, drinking water, gas chromatography, trihalomethane

HIGHLIGHTS

- Disinfection by-products in drinking water are neglected in African water utility companies.
- Potential health risks associated with disinfection by-products in drinking water.
- The headspace purge and trap coupled with a gas chromatography–mass spectrometry technique was used to quantify trihalomethanes.
- The WHO index was 0.4104, indicating no noncarcinogenic risk to human health in the study area.

GRAPHICAL ABSTRACT



INTRODUCTION

The availability and accessibility of affordable safe drinking water and sanitation services are not only basic human needs but also human rights. The human rights agenda necessitates that the water required for domestic consumption must be safe and free from micro-organisms, chemical, and radiological hazards that threaten human health (UN-Water 2019). Despite it being a basic human right, some populations do not have access to safe drinking water. In 2015, 2.1 billion people worldwide did not use a safely managed drinking water facility, whereas 844 million people still lacked a basic drinking water facility (WHO/UNICEF 2017). Also, the coverage of safe drinking water supply varies across regions: 94% for European countries and 24% for sub-Saharan African countries (UN-Water 2019).

Urban areas in developing countries are facing challenges in the provision of adequate and safe drinking water. This is attributed to more rapid population growth, and urbanization, which Africa has experienced over the past two decades in comparison to other regions (Bahri *et al.* 2016). The rapid population growth has led to an increase in water demand hence intermittent water supply. The intermittent water supply leads to reduced water supply pressures and also the water is highly susceptible to contamination (Vairavamoorthy 2007). Also, there is increased pollution of water sources attributed to poor solid waste and wastewater management practices in urban centers (Jain 2012). Furthermore, there is the challenge of climate change and this has led to extreme weather events such as floods that lead to damage to the water supply infrastructure, hence water loss (Bahri *et al.* 2016). Whereas droughts reduce the water levels leading to the deterioration of the water quality of water resources (Howard *et al.* 2016). Due to the polluted water sources, the water utility companies incur high treatment costs to provide safe water to the population.

Water utility companies are mandated to provide clean, safe potable drinking water to the population. To meet this requirement, disinfection by chlorination, ozonation, and ultraviolet light methods are used to kill pathogens including viruses and bacteria to ensure the water is free from disease-causing micro-organisms and, hence, fit for human consumption. Disinfection by chlorination is the most widely used method for the inactivation of pathogenic micro-organisms in water because it is readily available, easy to use, relatively low cost, and can form residual chlorine along the distribution system to avoid recontamination (USEPA 2011). This method provides hygienic quality drinking water.

Worldwide and in Africa, there is increasing pollution of surface water caused by point and non-point sources, which increases the organic matter load in the raw water abstracted for treatment (Fayiga *et al.* 2018). When the organic matter reacts with chlorine in the water in the form of hypochlorous acid and hypochlorite, there is the formation of unwanted disinfection by-products (DBPs), predominantly trihalomethane (THM) and haloacetic acids (HAA) (Durmishi *et al.* 2015). THM is categorized into four compounds, namely chloroform ($CHCl_3$), bromoform ($CHBr_3$), dibromochloromethane ($CHClBr_2$), and bromodichloromethane ($CHBrCl_2$). The chloroform takes the highest proportion of THMs in drinking water (Ristoiu *et al.* 2009).

THM formation is affected by the water source, bromide concentration, temperature, organic matter, pH, and residual chlorine (Guo *et al.* 2016). The natural organic matter (NOM) consists of chemical compounds categorized as hydrophilic parts comprised of carbohydrates, amino acids, and a hydrophobic part consisting of fulvic and humic acids. The hydrophobic part comprises 50–90% of total dissolved organic carbon (DOC) in surface water and is predominated by humic materials (Pérez *et al.* 2017). The humic materials are resistant to biodegradation, and instead, they react with chlorine to form increased levels of THM (Tak & Vellanki 2018). Chlorine reacts with aromatic compounds such as humic acids by electrophilic substitution. Electrophilic aromatic substitution is an organic reaction where an atom attached to an aromatic system that is hydrogen is substituted by an electrophile, hence the formation of THM (Bond *et al.* 2012).

The guideline values for chloroform, bromodichloromethane, dichloromethane, and bromoform in drinking water are 300, 60, 100, and 100 μg/L according to World Health Organization (WHO) guidelines (WHO 2017). For total THM, the sum of the ratio of the concentration of each individual compound to its respective guideline value should not exceed one in drinking water (WHO 2017). Disinfecting water by chlorination to protect public health from water-borne diseases poses health risks, such as carcinogens, and reproductive disorders in humans (Boorman *et al.* 1999; Richardson 2011; Pan *et al.* 2014). This is a concern that needs to be addressed by engineers, chemists, and policymakers in a way that elimination of pathogens is achieved and also no increase in the formation of DBPs in water supply systems that pose health risks (Boorman *et al.* 1999). Epidemiological studies have indicated health risks such as colon, rectal, and bladder cancers associated with disinfection by-products, whereas noncarcinogenic health risks included delayed pregnancies, stillbirth, and fetal loss among US women who had been exposed to DBPs (Villanueva *et al.* 2015).

Most developing countries have neglected the formation of the DBPs in drinking water and instead only focus on the microbiological WHO guidelines for drinking water (Durmishi *et al.* 2015). In Uganda, like any other developing country, surface water sources have become polluted. National Water and Sewerage Corporation (NWSC) is the public water utility company mandated to supply piped water and provide sewerage services across Uganda. Most municipal water supplies utilize disinfection by chlorination as part of the water treatment processes. Ggaba waterworks is the largest water supply for Kampala city supplying 226 m^3/day to approximately 2,551,299 million people and utilizes Lake Victoria as its water source (NWSC 2019).

Lake Victoria contains organic matter either from autochthonous sources where the organic matter is created in the interior of the water column from algae and phytoplankton activities, or from allochthonous sources where the organic matter is created from inputs getting into the water from outside the water body such as the discharge of industrial and municipal wastewater effluents, drainage, and runoff from agricultural fields near the lake shores (Deirmendjian *et al.* 2020). The allochthonous sources are dominated by hydrophobic contents; therefore, the formation of THM is inevitable (Tak & Vellanki 2018). According to the Lake Victoria Environmental Management Program (LVEMP), the organic matter levels in Lake Victoria were 7.5 mg/L (LVEMP 2005). In addition, a recent study carried out found out that the DOC was 23 mg/L in bays and 18.5 mg/L in open waters of Lake Victoria (Deirmendjian *et al.* 2020).

Based on the literature reviewed, no study on DBPs has been done at the Ggaba waterworks and along the distribution system. The study considered the evaluation of THM because they are the most common, regulated compounds, and are used as an indicator for the total DBP concentration (Whitaker *et al.* 2003). Therefore, the objectives of the study were to evaluate the THM in drinking water at the Ggaba waterworks, along the distribution system, to assess the relation between water quality parameters and the formation of THM, and estimate the potential health risks associated with THM in drinking water.

MATERIALS AND METHODS

Description of the study area

Kampala is the capital city of Uganda located in the central region. Ggaba waterworks is located in the Murchison Bay on the shores of Lake Victoria in Makindye Division approximately 11 km away from the central business district. Ggaba waterworks is the main water treatment plant supplying Kampala, Kira, and Mukono municipalities (Kalibbala *et al.* 2019).

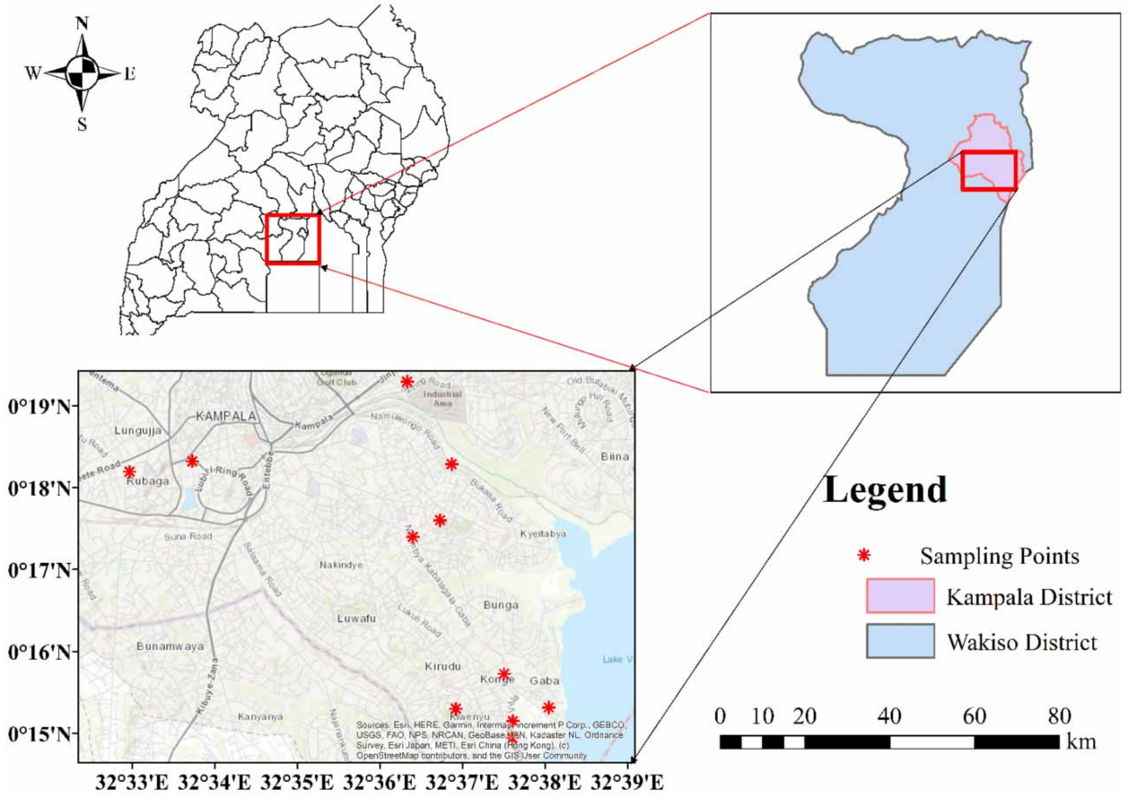


Figure 1 | A map of the study area and sampling points.

According to NWSC, Kampala water service coverage is divided into two zones: Kampala water eastern zone and Kampala water western zone for easy management. The western zone had approximately 18,500 households (NWSC 2019). Figure 1 shows the study area and the location of the sampling points.

Sampling strategy

Eleven sampling points were selected from the NWSC Kampala western zone's existing water sampling points. Three sampling points were selected from the treatment plant: raw water, filtered water before post chlorination, and the finished water after post chlorination, and eight were selected along the water distribution system. The THM concentration along the distribution system was categorized into three: points near the treatment plant P4 and P5 were 2 and 4 km, respectively, away from the plant, while intermediate points P6, P7, and P8 were 8, 10, and 13 km, respectively, away from the plant. The farther points P9, P10, and P11 were 14.6, 15.9, and 16.6 km, respectively, away from the treatment plant. These points were selected because they were easily accessible, tapping from the main pipeline therefore constant water supply. The samples were collected on Monday weekly between 8:30 am and 12:30 pm because at this time water production was at its peak. The sample collection lasted 12 weeks during January, February, and March 2021. A total number of 132 samples were collected during the study.

Sample collection procedures

The water samples for THM analysis were collected in duplicate in 40 mL glass vials with polypropylene screw caps and Teflon-faced septa. The vials were carefully washed, rinsed, and put in an oven at 100 °C for an hour before sampling. Sodium thiosulfate (3 mg) was added to the THM sample bottles to eliminate the further formation of THM. Before collecting samples from the sampling points, the taps were opened for about 3–5 min to ensure that the water was direct from the main distribution system. The vials were carefully filled to the brim without allowing air bubbles and sample overflow to avoid loss of THM due to their volatility. After sampling, the vials were put in a cooler box and kept at 4 °C as they were transported from the field to the laboratory. The samples were kept at the same temperature until analysis which was done within 48 h. The water samples for the determination of other water parameters such as total organic carbon, bromide, and UV_{254} were collected in duplicate in 250 mL glass bottles.

Analytical methods

THM present in water was measured following USEPA method 524.2. The automatic headspace purge and trap system was coupled to a gas chromatography–mass spectrometry detector. The purge and trap technique was used in the extraction of THM because it does not require the use of expensive solvents, and it is a fast technique. The gas chromatography ensures compounds in the sample are chromatographically separated depending on their volatility and polarity, while the mass spectrometer confirms the occurrence of the compounds according to their mass fragments. The GC capillary column length of 15 m, internal diameter of 0.25 mm, and 1.0 μm film thickness were used in the chromatographic separation. The GC temperature profile was set to a start temperature of 60 °C and held for 2 min and 30 s, with a temperature increase to 100 °C at a rate of 10 °C/min, a final temperature increase to 180 °C at a rate of 26 °C/min. Injector and detector temperatures were 230 and 250 °C, respectively. The carrier gas, nitrogen, was set in constant flow mode at 16.7 mL/s to the GC column. The whole process took a maximum of 10 min.

A stock standard containing 400 mg/L of each of the individual THM, chloroform, bromodichloromethane, bromoform, and dibromochloromethane Spexcertiprep brand was used. Calibration standards of 10, 20, 50, and 100 μg/L were prepared and analyzed to find out the response of the instrument to the known concentrations. The chromatograms were recorded on the computer, and the peak areas of individual THM are proportional to the individual THM concentrations in μg/L (APHA 2012). The retention times were 1 min 6 s, 1 min 41 s, 2 min 47 s, and 4 min 20 s for chloroform, bromodichloromethane, dibromochloromethane, and bromoform, respectively. The four-point calibration curves were plotted over the well-known concentration range. The linear regression of peak area versus concentration for the individual THM gave a good fit R^2 of 0.9978, 0.9971, 0.9988, and 0.9935 for chloroform, bromodichloromethane, dibromochloromethane, and bromoform, respectively. The molecular ions (M^+) for chloroform, bromoform, dibromochloromethane, and bromodichloromethane were detected at mass/charge of 83, 173, 129, and 83 following a 70-electron volt high-energy ionization in a mass spectrometer. A set of eight samples of a volume of 20 mL were spiked with 50 μL from a 400-mg/L standard concentration. The unspiked and spiked samples were run, and a relative standard deviation of 3.5% for chloroform and 7.9% for bromodichloromethane for spiked samples indicated a high precision since relative standard deviations were <20%. Also, the rate of recovery for chloroform ranged from 95 to 108%, while for bromodichloromethane recovery ranged from 97 to 103%, indicating a high accuracy level for the technique. The detection limit of the technique was 0.02 μg/L.

Determination of water quality parameters in drinking water

The TOC measurements were done in the laboratory following Standard Method 5310C (APHA 2012). The TOC was analyzed using a laboratory total organic carbon analyzer model Sievers M5310C. The ultraviolet absorbance at wavelength 254 nm (UV_{254}) measurement was carried out in the laboratory using standard method 5910B (APHA 2012). Ultraviolet absorbance was measured at 254 nm wavelength using a DR spectrophotometer model (Cecil-400) with a 10 mm long quartz cell. Organic free water UV_{254} absorbance value was used as a reference. The bromide concentration was measured in the laboratory using the phenol red colorimetric method 4500C (APHA 2012). The water pH, temperature, and residual chlorine were measured in the field because the transportation and storage affect their properties. The residual chlorine was measured using a potable palintest chlorometer while utilizing the diethyl phenyl diamine DPD tablets. The water temperature and pH were measured using a portable pH meter following the Standard Method (APHA 2012). The pH meter was calibrated before use with standard pH 4.0, 7.0, and 10.0 buffer solutions. The pH meter also measured the water temperature.

Potential health risk estimation for THM

The potential health risk associated with THM was estimated using two models as detailed below.

WHO Index (I_{WHO})

The WHO index is an estimation for additive toxicity for THM. The index was used to estimate the toxic risk associated with chlorinated water. For compliance, the index should be less than one (WHO 2017). The WHO index was computed using the following equation:

$$I_{\mathrm{WHO}} = \frac{C_{\mathrm{CF}}}{GV_{\mathrm{CF}}} + \frac{C_{\mathrm{BDCM}}}{GV_{\mathrm{BDCM}}} + \frac{C_{\mathrm{BF}}}{GV_{\mathrm{BF}}} + \frac{C_{\mathrm{DBCM}}}{GV_{\mathrm{DBCM}}} \leq 1 \tag{1}$$

where C is the concentration of chloroform, bromodichloromethane, bromoform, and dibromochloromethane measured according to WHO guideline values for THM in drinking water (chloroform 300 μg/L, bromodichloromethane 60 μg/L, dibromochloromethane 100 μg/L, bromoform 100 μg/L) (WHO 2017).

USEPA risk estimation method

The toxic risk is defined as the hazard quotient (HQ) (Ferreira & Cunha 2012). The HQ was computed using the following equation:

$$\mathrm{HQ} = \frac{\text{Total amount ingested}}{\mathrm{BW} \times \mathrm{ET} \times \mathrm{RfD}} \tag{2}$$

where BW is the body weight, kg; ET is the exposure time, days/year; and RfD is the reference dose, mg/kg/day. The reference doses were obtained from toxicological studies of exposure that demonstrate critical effects as stipulated by the Integrated Risk Information System database (Ferreira & Cunha 2012).

Carcinogenic risk estimation

Carcinogenic risk assessment commonly considers that the risk is proportional to the total lifetime dose. The exposure system of measurement used to estimate the carcinogenic risk assessment was the lifetime average daily dose (LADD) (Ewaid *et al.* 2018). LADD is an estimate of the daily intake of a carcinogenic agent throughout the whole life of an individual. The cancer slope factor is the gradient of the line of the dose-response curve derived from laboratory toxicological studies, and values for each THM were obtained from the USEPA IRIS database (Ferreira & Cunha 2012). There are several ways in which humans are exposed to carcinogens, that is through ingestion, skin contact, and inhalation. The following computations according to USEPA guidelines were used to compute the carcinogenic risks for different exposure routes (Pardakhti *et al.* 2011).

Carcinogenic risk through ingestion

The carcinogenic risk through ingestion of THM in drinking water was computed using the following equations:

$$\text{THM carcinogenic risk by ingestion} = \mathrm{LADD}_{\text{Ingestion}} \times \mathrm{CSF}_{\text{Ingestion}} \tag{3}$$

$$\mathrm{LADD}_{\text{Ingestion}} = \frac{\text{THM concentration} \times \mathrm{ER} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{4}$$

where ER is the exposure rate, water consumption rate liters/day, EF is the exposure frequency days of the year for water consumption, ED is the exposure duration that is equivalent to the life expectancy of the population, BW is the body weight (kg), and AT is the average exposure time (days/year).

Carcinogenic risk through skin contact

The carcinogenic risk through skin contact was computed using the following equations:

$$\text{THM carcinogenic risk by skin contact} = \mathrm{LADD}_{\text{skin contact}} \times \mathrm{CSF}_{\text{skin contact}} \tag{5}$$

$$\mathrm{LADD}_{\text{skin contact}} = \frac{\text{THM concentration} \times \mathrm{BSA} \times \mathrm{PC} \times \mathrm{ED} \times \mathrm{EF} \times \mathrm{ET}}{\mathrm{BW} \times \mathrm{AT}} \tag{6}$$

where BSA is the body surface area, m^2, PC is the permeability coefficient, cm/h, and ET is the exposure time in the shower, min/day. The body surface area for the Kampala western zone was computed according to the following equation (Reading & Freeman 2005):

$$\mathrm{BSA} = \frac{1}{6}\,(\text{Body weight } \times \text{Height})^{0.5} \tag{7}$$

Carcinogenic risk through inhalation

The cancer risk through inhalation was considered for chloroform (USEPA 1991). The risk was computed using the following equations:

$$\text{THM carcinogenic risk inhalation} = \text{LADD}_{\text{inhalation}} \times \text{CSF}_{\text{inhalation}} \quad (8)$$

$$\text{LADD}_{\text{skin contact}} = \frac{\text{Chloroform concentration} \times \text{AA} \times \text{VF} \times \text{ED} \times \text{EF} \times \text{ET}}{\text{BW} \times \text{AT}} \quad (9)$$

where AA is the aspirated air, m^3/day, and VF is the volatilization factor for chloroform l/m^3. The total carcinogenic risk for all the three exposure routes was computed by summing up individual exposure risks. According to the USEPA, the cancer risk is divided into four categories: if $CR<1\times10^{-6}$ the risk is negligible, $1\times10^{-6}<CR<5.1\times10^{-5}$ acceptable low risk, $5.1\times10^{-5}<CR<1\times10^{-4}$ acceptable high risk, and if $CR>1\times10^{-4}$ then the risk is unacceptable (Legay *et al.* 2011). Exposure factors required in Equations (2), (4), (6), (7), and (9) such as aspirated air, permeability coefficient, and volatilization factor were obtained from the existing USEPA standard data, as shown in Table 1.

Also, to use the USEPA model for estimating the potential health risk, the cancer slope factors and the reference doses for the compounds of chloroform and bromodichloromethane were obtained from the online integrated risk information system (IRIS), as shown in Table 2.

Data analysis

Quantitative data collected was analyzed using Microsoft Office Excel and Minitab computer package version 20 for Windows. The data were summarized and tabulated using Microsoft Excel where the mean and standard errors for the data were computed. Also, a one-way analysis of variance (ANOVA) was used to determine whether the mean differences of total THM for the different points were significant or not. Minitab was used to determine the Pearson correlation coefficients.

RESULTS AND DISCUSSION

THM concentration at Ggaba water treatment plant

The results of the THM measurement at the Ggaba waterworks are summarized in Table 3.

Table 1 | Exposure factors for the exposure assessment

Exposure factor	Notation	Unit	Value	Reference
THM Conc. in water	CF BDCM	μg/L	CF: 19.91 BDCM: 7.86	This study
Exposure rate	ER	L/day	2	USEPA (1997)
Exposure frequency	EF	days/year	365	Abbas *et al.* (2015)
Exposure duration	ED	year	Female: 64.5 Male: 62.8	UBOS (2013)
Average exposure time	AT	days/year	Female: 64.5×365 Male: 62.8×365	Lee *et al.* (2004) Abbas *et al.* (2015)
Bodyweight	BW	kg	Female: 67.1 Male: 70.5	This study
Surface area	SA	m^2	Female: 1.69 Male: 1.84	This study
Exposure time	ET	min/day	35	USEPA (1997)
Aspirated air	AA	m^3/day	20	Semerjian & Dennis (2008)
Volatilization factor for chloroform	VF	L/m^3	0.5	Semerjian & Dennis (2008)
Permeability coefficient	PC	cm/h	CF-0.00683 BDCM-0.00402	IRIS (2012)

Table 2 | Carcinogenic slope factors and reference doses for the individual THM

Chemical compound	Cancer groups	Carcinogenic slope factor (CSF) (mg/kg day) Oral/dermal	Carcinogenic slope factor (CSF) (mg/kg day) Inhalation	Reference dose (RfD)	Reference
Chloroform	B1	3.1×10^{-2}	8.05×10^{-5}	0.01	IRIS (2012)
Bromodichloromethane	B2	6.2×10^{-2}		0.02	IRIS (2012)

Table 3 | Trihalomethanes concentration along the Ggaba water treatment plant

Parameter	Chloroform (μg/L)			Bromodichloromethane (μg/L)			Total trihalomethanes (μg/L)		
Point	P1	P2	P3	P1	P2	P3	P1	P2	P3
Mean	ND	10.53	19.86	ND	4.18	4.59	ND	14.72	24.45
±SE		0.88	2.34		0.73	0.59		1.53	2.39
Min.		3.99	11.87		0.69	0.78		4.68	14.96
Max.		14.42	33.98		8.99	9.43		23.41	38.65
NWSC standard									**80.00**
WHO Guideline		**200**	**200**		**60**	**60**			**100**
USEPA standard									**80**

Bold values are standard values and guideline values for the respective bodies.

The mean chloroform, bromodichloromethane, and total THM concentration obtained at the treatment plant were all within the NWSC, WHO, and USEPA acceptable drinking water guidelines and standards, respectively. The data were normally distributed; therefore, a single factor analysis of variance (ANOVA) statistical test was carried out to determine whether the differences between mean concentrations of TTHM for points P2 and P3 were statistically significant or not. The one-way ANOVA test indicated that the differences between mean concentrations of TTHM at points P2 and P3 were statistically significant ($p\leq0.05$).

There was no THM in the raw water because for THM to form, there should be precursors of NOM in presence of chlorine. However, the raw water received at the plant did not have chlorine, hence the absence of THM in raw water samples. The filtered water had THM because of the pre-chlorination process done at the treatment plant. Pre-chlorination is done to reduce the algae that are in the raw water source. The final treated water showed an increase in the total THM because of the post chlorination done at the water treatment plant to ensure pathogens are killed and also maintain a given concentration of residual chlorine along the distribution system.

Generally, the trend of individual THM found at Ggaba waterworks showed that chloroform concentration levels were higher than the bromodichloromethane concentration levels. The chloroform concentration at the waterworks contributed 81.2% to the TTHM compared to 18.8% contributed by bromodichloromethane. This result has been supported by a study carried out in Ethiopia where it was found out that the chloroform concentrations were higher than other THM in the Hossana water supply system (Zelelew *et al.* 2018). Also, another study carried out in Cairo Egypt found higher chloroform concentration levels compared to other THM in the water (Souaya *et al.* 2014). The low levels of bromodichloromethane were attributed to low levels of bromide concentration in the water source.

THM concentration along the water distribution system

Table 4 summarizes the results of THM measurement for near, intermediate, and far points along the water distribution system.

The mean chloroform, bromodichloromethane, and total THM concentration obtained along the water distribution system were all within the NWSC, WHO, and USEPA acceptable drinking water guideline and standards, respectively. The mean total THM concentration increased in order P11>P10>P9>P8>P7>P6>P5>P4. This is because the longer the distance, the contact time between residual chlorine and the organics in water increases hence increasing the formation of total

Table 4 | Trihalomethane concentration along the water distribution system

		Near points		Intermediate points			Far points		
Parameter/Point		**P4**	**P5**	**P6**	**P7**	**P8**	**P9**	**P10**	**P11**
Chloroform (μg/L)	Mean	16.16	17.77	19.17	19.21	20.76	20.8	23.07	22.39
	SE±	0.59	0.56	1.93	1.2	1.25	1.36	1.47	2.05
	Min.	12.49	14.85	11.23	12.31	11.45	11.23	14.8	13.12
	Max.	19.87	21.79	28.12	24.79	27.43	26.88	35.65	39.3
BDCM (μg/L)	Mean	5.68	5.7	7.31	7.58	7.74	9.59	8.79	10.5
	SE±	0.51	0.43	1.15	0.84	0.98	1.1	0.68	1.0
	Min.	2.78	2.95	2.38	0.89	1.85	2.79	4.53	4.99
	Max.	9.03	7.65	13.56	10.61	11.6	15.49	11.83	14.42
TTHM (μg/L)	Mean	21.84	23.47	26.49	26.79	28.5	30.38	31.86	32.89
	SE±	0.95	0.88	2.81	1.64	2.07	2.21	1.77	2.93
	Min.	18.23	17.8	16.45	19.76	15.77	16.57	19.33	18.4
	Max.	28.9	29.44	41.18	35.4	39.03	39.75	41.87	53.72

THM (Saidan *et al.* 2013; Zelelew *et al.* 2018). The one-way ANOVA test indicated that the differences between mean concentrations of TTHM at the plant, near, intermediate points, and far points along the water distribution system were all statistically significant ($p \leq 0.05$).

Relationship between water quality parameters and THM formation

A Pearson correlation method was performed and Table 5 illustrates the correlation coefficients r between the total THM and other drinking water quality parameters.

Residual chlorine and THM

A weak, negative but significant correlation of $r = -0.332$, $p = 0.001 < 0.05$ was obtained between total THM and residual chlorine. Generally, as the residual chlorine content decreased, the concentration of THM increased. This is because the presence of chlorine in water dissociates into hypochlorous acid and hypochlorite ion species which are pH-dependent. Hypochlorous acid is dominant whenever conditions are acidic, whereas hypochlorite ion dominates when the conditions are alkaline. During the study, pH values in drinking water ranged from 6.6 to 7.5. Thus, hypochlorous acid was more dominant than hypochlorite ion. Therefore, the hypochlorous acid reacted with the organic matter to form THM. In this case, hypochlorous acid is consumed to generate THM, hence a reduction in residual chlorine. Several studies have been conducted and have reported the same trend; the reduced residual chlorine was associated with an increased THM formation (Abdullah *et al.* 2003; Kowalska *et al.* 2007; Ye *et al.* 2009; Ramavandi *et al.* 2015). Also, due to other factors such as chlorine decay, quantities of chlorine doses could affect the relationship between residual chlorine and THM concentration. This is supported

Table 5 | Pearson's correlation matrix for the water quality parameters

Parameter	**Residual chlorine (mg/L)**	**Temp. (°C)**	**pH**	**TOC (mg/L)**	**UV_{254} (cm^{-1})**	**Bromide (mg/L)**	**Chloroform (μg/L)**	**BDCM (μg/L)**
Temperature (° C)	−0.256							
pH	−0.241	0.149						
TOC (mg/L)	−0.422	0.28	0.3					
UV_{254} (cm^{-1})	−0.425	0.248	0.1	0.336				
Bromide (mg/L)	−0.238	0.833	0.037	0.186	0.308			
Chloroform (μg/L)	−0.294	0.863[a]	0.089	0.305	0.234	0.841		
BDCM (μg/L)	−0.329	0.8[a]	0.059	0.283	0.341	0.984[a]	0.725	
TTHM (μg/L)	−0.332	0.905[a]	0.097	0.324	0.280	0.873[a]	0.95[a]	0.883[a]

Correlations at 95% confidence level.

[a] Strong positive correlations.

from the literature by other studies that have found out, contrary to our study, a positive correlation between total THM and residual chlorine (Zelelew *et al.* 2018; Nadali *et al.* 2019). This is explained by increased chlorine doses leading to increased available residual chlorine hence increased THM formation (Saidan *et al.* 2013). Thus, water treatment plants struggling with high THM values should ensure that they either remove most of the organic matter during coagulation or lower the chlorine dose to a prior determined threshold where THM generated conforms to the standards but without compromising on the need for residual chlorine.

Total organic carbon and THM

Using a Pearson correlation method, a weak, positive but significant correlation $r=0.324$, $p=0.001<0.05$ was obtained between total THM and TOC, as seen in Table 5. The trend showed that at points with high total organic carbon, there was an increase in the total THM formed. This implies that the NOM present in the water favors the formation of THM. According to Abdullah *et al.* (2003), the hydrophobic part of the NOM is majorly responsible for the THM formation since the part readily reacts with chlorine. Also, several studies have been carried out which found out that THM and total organic matter have a low, positive, and significant correlation (Abdullah *et al.* 2003; Ye *et al.* 2009; Saidan *et al.* 2013; Souaya *et al.* 2015). Contrary to this study, other studies have been carried out and found that TOC and THM have a negative correlation explanation as not all the organic compounds present in water lead to the formation of THM. Furthermore, the total organic carbon measurements do not distinguish between the various chemical compounds comprising the DBP precursor (Mohammadi *et al.* 2015; Zelelew *et al.* 2018; Nadali *et al.* 2019).

pH and THM

According to the Pearson correlation method, a very weak positive but non-significant correlation $r=0.097$, $p=0.348>0.05$ was obtained between total THM and pH for the study. The pH values were in a narrow range from 6.6 to 7.5. This is because the pH is strictly maintained within the acceptable limits, hence the very weak correlation between pH and THM formation. However, previous studies have found out that as the pH levels increased from 6 to 9, the total THM formed increases (Ye *et al.* 2009; Saidan *et al.* 2013; Souaya *et al.* 2015; Zelelew *et al.* 2018). This is because higher pH levels favor base-catalyzed hydrolysis of the halogenated group, of which THM are a part (Bond *et al.* 2012). In addition, according to Bond *et al.* (2012), when the pH levels are 5 and below being acidic conditions, studies have indicated a decrease in THM formation.

Bromide concentration and THM

A strong positive but significant correlation $r=0.873$, $p=0.00<0.05$ was obtained between total THM and bromide concentration. The study showed an increase in the bromide concentrations led to increased total THM, which is contributed by an increase in the formation of bromodichloromethane. This is explained by the presence of bromide in water which is oxidized to bromine that reacts with NOM to form brominated THM (Kujlu *et al.* 2020). Several researchers have found out that a slight increase in bromide concentrations leads to an increase in total THM (Ristoiu *et al.* 2009; Ye *et al.* 2009; Souaya *et al.* 2015).

Ultraviolet absorbance at 254 nm and THM

A weak, positive but significant correlation $r=0.280$, $p=0.006<0.05$ was obtained between total THM and ultraviolet absorbance at 254 nm. Generally, high UV_{254} values led to an increase in the formation of THM. This is explained by a high UV_{254} which means there is a high concentration of aromatic material in the water which consists of the primary sites that react with chlorine to form DBPs (Stéphanie & Caetano 2020). According to a study carried out in China on factors influencing DBPs formation in drinking water, the increased UV_{254} levels led to the increased formation of THM, whereas whenever the UV_{254} levels were low, the THM formation was low (Ye *et al.* 2009).

Temperature and THM

A strong positive and significant correlation $r=0.905$, $p=0.00<0.05$ was obtained between total THM and temperature. Generally, the trend on the plot from the study showed that as temperatures increased, the total THM formed also increased. This is explained by an increase in temperature that quickens the reaction rate between organic matter present in water and chlorine (Ramavandi *et al.* 2015). Several studies have been conducted to investigate factors affecting THM formation in drinking water and results indicated that increased temperatures led to an increase in THM formed (Saidan *et al.* 2013; Ramavandi *et al.* 2015; Zelelew *et al.* 2018).

Evaluation of potential health risks associated with THM

The exposure and assessment of the cancer risk were calculated based on the mean concentration of the individual THM found in the water; chloroform 19.92 μg/l and bromodichloromethane 7.86 μg/l measured during the study.

WHO index for noncarcinogenic risks for THM

The WHO index for additive toxicity was calculated using Equation (1). A WHO index of 0.4, which was less than one, was obtained. Due to this, the additive toxicity of THM levels in the Kampala water western zone is compliant with the WHO guideline. According to the WHO index, the value showed that the levels of THM in water do not pose any noncarcinogenic risk to the residents in the Kampala western zone. Previous studies have been done and the WHO index for additive toxicity was less than unity, meaning that the concentrations did not pose any adverse toxic health impacts (Semerjian & Dennis 2008; Salih & Al-azzawi 2016; Ewaid *et al.* 2018).

USEPA carcinogenic risk assessment

The sample size of 376 people was used in the survey which provided a representative body weight and height for Kampala water western zone population. The average body weights obtained for males and females were 70.6 and 67.1 kg, respectively, while the average heights were 164 and 158.8 cm for males and females, respectively. The surface body area was computed using Equation (7), and this resulted in 1.84 and 1.69 m^2 surface area for males and females, respectively. The results obtained in the study are supported by the USEPA (1997) standards of 70 kg for an adult body weight, and body surface areas of 1.9 and 1.69 for males and females, respectively. To carry out the USEPA carcinogenic risk assessment, various exposure factors were obtained from Tables 1 and 2. Also, according to the Uganda Bureau of Statistics, the average life expectancy for Ugandans was 62.8 and 64.5 years for males and females, respectively. These factors were used in the computation of the carcinogenic risks.

HQ evaluation for THM

The HQ for individual THM was computed using Equation (2); the HQ chloroform was 0.1 which is less than one, while the HQ bromodichloromethane was 0.01 which is less than one. Therefore, the THM concentrations found in the distribution system do not pose any adverse developmental and noncarcinogenic risks for CF and BDCM to the Kampala water western zone residents during the study period according to USEPA guidelines. Several studies have used the HQ to evaluate the risk and results showed that there were no noncarcinogenic risks (Viana & Cavalcante 2009; Karim *et al.* 2013; Abdullah 2014; Ewaid *et al.* 2018).

Different exposure route evaluations of lifetime cancer risks for THM

The computation for the lifetime carcinogenic risk for THM was obtained using exposure factors in Tables 1 and 2 substituted in Equations (3)–(9). Table 6 summarizes the carcinogenic risks through oral, dermal, and inhalation routes for females and males computed for the study population.

Oral route evaluations of lifetime carcinogenic risk for THM

The cumulative carcinogenic risk through the oral route for the total THM as seen in Table 6 lies within the USEPA acceptable low-risk range of $1\times10^{-6} < \text{carcinogenic risk} < 5.1\times10^{-5}$. The oral carcinogenic risk is within the acceptable low-risk range which is the same as a study carried out in Pakistan (Siddique *et al.* 2015). Chloroform and bromodichloromethane contributed 71.6 and 28.3%, respectively, to carcinogenic risk through the oral route. A study carried out in India showed that chloroform had the highest percentage contribution of 56% to total oral carcinogenic risks (Mishra *et al.* 2014). Generally, the oral carcinogenic risk was higher in females than in males; this was attributed to the high life expectancy of Ugandan females compared to the males. Chloroform presents a great carcinogenic risk though within the acceptable low-risk range compared to bromodichloromethane because the concentration of chloroform was much higher than for bromodichloromethane in the water samples during the study.

Dermal route evaluations of lifetime carcinogenic risk for THM

The cumulative carcinogenic risk through the dermal route for the total THM as seen in Table 6 lies within the USEPA acceptable low-risk range of $1\times10^{-6}<\text{carcinogenic risk}<5.1\times10^{-5}$. A study carried out in Abadan gave the same range of acceptable low risk for the dermal route of THM in water (Kujlu *et al.* 2020). Chloroform and bromodichloromethane

Table 6 | Lifetime carcinogenic risks associated with trihalomethane in water

Chemical compound	Oral exposure risk	Dermal exposure risk	Inhalation exposure risk	Total risk
	Female			
Chloroform	1.82×10^{-5}	7.37×10^{-6}	8.29×10^{-6}	3.39×10^{-5}
Bromodichloromethane	7.20×10^{-6}	1.71×10^{-6}		8.91×10^{-6}
Total Risk	2.54×10^{-5}	9.08×10^{-6}	8.29×10^{-6}	4.28×10^{-5}
	Male			
Chloroform	1.75×10^{-5}	8.12×10^{-6}	7.96×10^{-6}	3.36×10^{-5}
Bromodichloromethane	6.91×10^{-6}	1.89×10^{-6}		8.80×10^{-6}
Total Risk	2.44×10^{-5}	1×10^{-5}	7.96×10^{-6}	4.24×10^{-5}

contributed 81.2 and 18.8%, respectively, to carcinogenic risk through the dermal route. Chloroform presents a great risk though within the acceptable low-risk range compared to bromodichloromethane because the concentration of chloroform was much higher than for bromodichloromethane in the water samples during the study.

Inhalation route evaluations of lifetime carcinogenic risk for THM

The estimation of THM carcinogenic risk through the inhalation route was conducted for only chloroform because it is highly volatile (Siddique *et al.* 2015). The carcinogenic risk through the inhalation route as seen in Table 6 lies within the USEPA acceptable low-risk range of $1\times10^{-6}<\text{carcinogenic risk}<5.1\times10^{-5}$. This is explained by the generally low concentration of chloroform during the study. However, compared to previous studies done in Abadan and India, the inhalation carcinogenic risk of chloroform was found to be in an acceptable high-risk range (Mishra *et al.* 2014; Kujlu *et al.* 2020).

Total carcinogenic risk for THM

The results indicated in Table 6 that the total carcinogenic risk lies within the USEPA acceptable low-risk range of $1\times10^{-6}<\text{carcinogenic risk}<5.1\times10^{-5}$ and risk through the oral route>dermal route>inhalation route for both males and females. Previous studies carried out found the carcinogenic risk of THM through the oral route>dermal route>inhalation route (Lee *et al.* 2004; Semerjian & Dennis 2008; Ewaid *et al.* 2018).

CONCLUSION

During the study, only chloroform and bromodichloromethane species were present at the water treatment plant and the water distribution system. Chloroform ranged from 11.9 to 33.9 μg/L and 11.2 to 39.3 μg/L at the treatment plant and along the distribution system, respectively, whereas bromodichloromethane ranged from 0.8 to 9.4 μg/L and 0.9 to 9.15.5 μg/L at the treatment plant and along the distribution system, respectively. The concentrations of THM found in the Kampala water western zone were within NWSC (80 μg/L), WHO (300 μg/L for chloroform and 60 μg/L for bromodichloromethane), and USEPA (80 μg/L) guidelines.

A Pearson correlation was run to find the relation between water parameters TOC, UV_{254}, bromide concentration, temperature, pH, residual chlorine, and formation of THM. The correlation showed $r=0.324$ and $p=0.001$ for TOC, $r=0.28$ and $p=0.006$ for UV_{254}, $r=0.873$ and $p=0.00$ for bromide concentration, $r=0.905$ and $p=0.00$ for temperature, $r=0.097$ and $p=0.348$ for pH, and $r=-0.332$ and $p=0.001$ for residual chlorine. The TOC and UV_{254} had a weak positive but significant correlation; however, the bromide concentration and the temperature had a strong positive and significant correlation with THM formation. In addition, pH had a very weak positive but non-significant correlation, while the residual chlorine had a weak negative but significant correlation with THM formation.

Potential health risks associated with THM were evaluated using the WHO index and the USEPA model. The WHO index was 0.4 which was less than unity, indicating no noncarcinogenic risk to human health in the study area. Also, the HQ was less than unity, indicating no adverse health effects due to exposure to THM in the Kampala western zone. The human lifetime carcinogenic risks of THM due to oral ingestion, dermal, and inhalation were 2.5×10^{-5}, 9.1×10^{-6}, and 8.3×10^{-6} for females and 2.4×10^{-5}, 1×10^{-5}, and 7.9×10^{-6} for males. The values were within the USEPA acceptable low-risk range of

$1\times10^{-6}<CR<5.1\times10^{-5}$. Therefore, the residents in the Kampala Water western zone are consuming water that has a low acceptable carcinogenic risk according to USEPA standards.

RECOMMENDATIONS

There is a need for a continuous monitoring program for THM at the Ggaba water treatment plant and also along the distribution system. This is necessary to balance the use of chlorine to kill pathogens while putting into consideration the minimization of the formation of THM.

Chlorine as a disinfectant should continue to be used but the team at the Ggaba water treatment plant should ensure that all the treatment units preceding disinfection are operating effectively such that the total organic matter is reduced. Thus, this will lead to a further reduction in the formation of THM formed.

A study to monitor the other chlorination by-products in chlorinated drinking water, such as halo acetic acids, halo aldehydes, and halo-ketones, at the Ggaba water treatment plant and other water treatment plants supplying other districts should be done. Additionally, a study needs to be carried out including the seasonal variations and the distribution of THM concentrations at the plant and along the water distribution system.

There is a need for the National Environmental Management Authority to enforce the regulations on effluent discharges from industrial, wastewater treatment plants into the water sources. This will in turn reduce the organics getting into the raw water sources.

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DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

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