

ANALYSIS OF TRIHALOMETHANES AND HALOACETIC ACIDS IN TAP WATER OF NAIROBI CITY, KENYA

Osoro, V*

Oduor, T**

Kiptoo, J*

Mavura, W*

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ABSTRACT

Over 1.1 billion people in the world lack access to portable water. Diarrheal and other waterborne diseases cause an estimated 2.2 million deaths per year. Chlorination is a proven water treatment method for municipal water. Concerns have been raised about the potential long term health effects of disinfection by-products to users of chlorinated water. This study investigated the production of trihalomethanes (THMs) and Haloacetic acids (HAAs) in chlorinated tap water from Nairobi City, Kenya. The study was done using Gas Chromatography-Electron Capture (GC-ECD). Tap water samples were collected from ten locations around the city for analysis of THMs and HAAs. The study also investigated the effects of pH, Temperature, Colour, Turbidity, Ultra Violet Absorption and organic carbon. Data obtained was subjected to Microsoft Excel and analysis of variance (ANOVA). Some samples showed significant amount of Chloroform: ranging between 7.4 μ g/L and 13.06 μ g/L. Other samples showed traces of dibromochloromethane; 1.7 μ g/L and 6.6 μ g/L. Results from this study are meant to highlight the problems of chlorine contamination and the need for proper water treatment.

* Chemistry Department, Jomo Kenyatta University of Agriculture and Technology, P.O BOX 62000-00200, Nairobi, Kenya.

** Chemistry Department, Kenya Bureau of Standards, P.O BOX 54974-00200, Nairobi, Kenya.

1.0 INTRODUCTION

Access to safe drinking water has improved steadily and substantially over the last decades in almost every part of the world (Lomborg and Björn, 2001). However, some observers have estimated that by 2025 more than half of the world population will be facing water-borne diseases (Kulshreshtha, 1998). Water plays an important role in the world economy, as it functions as a solvent for a wide variety of chemical substances and facilitates industrial cooling and transportation. Approximately 70% of freshwater is consumed by agriculture (Baroni *et al.*, 2007).

According to a 2007 World Health Organization report, 1.1 billion people lack access to an improved drinking water supply, 88% of the 4 billion annual cases of diarrheal disease are attributed to unsafe water and inadequate sanitation and hygiene, and 1.8 million people die from diarrheal diseases each year. In addition, waterborne diarrheal diseases lead to decreased food intake and nutrient absorption, malnutrition, reduced resistance to infection (Baqui *et al.*, 1993), and impaired physical growth and cognitive development (Guerrant *et al.*, 1999). The WHO estimates that 94% of these diarrheal cases are preventable through modifications to the environment, including access to safe water (Lomborg and Björn, 2001). Chlorination was first used for disinfection of public water supplies in the early 1900s, and is one factor that contributed to dramatic reductions in waterborne disease in cities in the United States (Cutler and Miller, 2005).

Chlorine is usually added to water in the form of chlorine gas, or as a sodium or calcium hypochlorite solution. Chlorine itself hydrolyzes quickly to hydrochloric acid and hypochlorous acid. These compounds may dissociate, dependent on the pH of the solution. The overall reaction scheme is given in Figure 1.

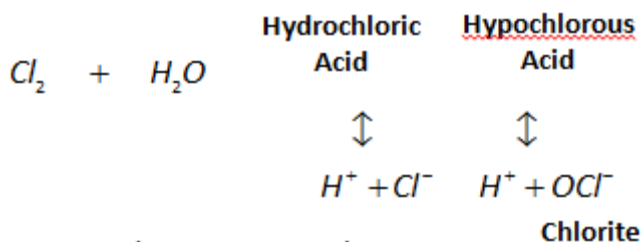
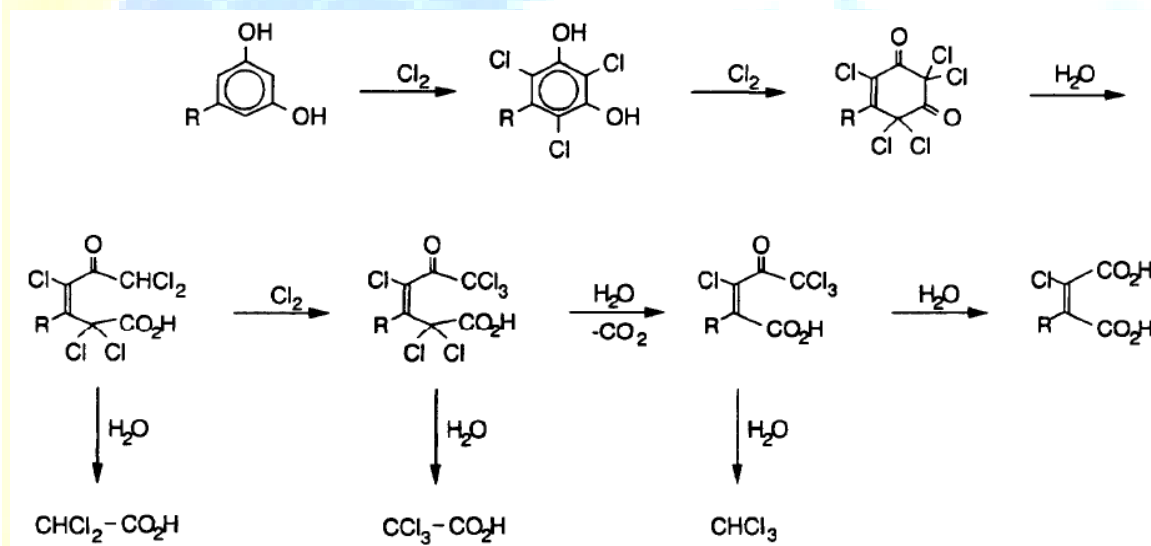


Figure 1: Overall reaction scheme

Humic material is a complex collection of different, sometimes polymeric, substances such as lignin, carbohydrates, proteins and fatty acids. It originates from deteriorated plants and other organic material and was one of the first sources associated with the formation of trihalomethanes. A considerable amount of research has since then been put in unveiling the general composition of humic material and in determining its susceptibility to chlorination reactions. It was found that especially phenolic compounds are susceptible to chlorination via a ring rupture mechanism (Reckhow and Singer, 1990).

The haloform reaction is base-catalyzed and, as such, the rate is dependent on the pH of the reaction medium. The base-catalyzed reaction removes a proton from the carbon double bond causing the formation of the enolate ion. This is the rate determining step and the enolate ion will subsequently undergo chlorine substitution at the carbon in the alpha position (the carbon next to the carbonyl carbon).



The resulting trichloromethyl ketone then hydrolyzes, causing the cleavage of the carbonyl carbon and alpha carbon bond and yielding chloroform and a carboxylic acid.

2.0 MATERIALS AND METHODS

2.1 The Study Area (Location and Environment)

Nairobi occupies an area of about 700 square kilometers at the south-eastern end of Kenya's agricultural heartland. At 1 600 to 1 850 m above sea level, it enjoys warm temperatures year

round (CBS 2001, Mitullah 2003). The Nairobi, Ngong, and Mathare rivers traverse numerous neighborhoods and the indigenous Karura forest still spreads over parts of northern Nairobi. The Ngong hills are close by in the west, Mount Kenya rises further away in the north, and Mount Kilimanjaro emerges from the plains in Tanzania to the south-east. The bulk of water supply for Nairobi comes from Thika, Sasumua and Ruiru Dams, as well as the Kikuyu Springs. Over time water supply for the city has failed to meet demand.

Most residents in Nairobi city get their portable water from Kabete water production plant, whereas residents along Thika road get from Ngethu water production plant. Jomo Kenyatta University of Agriculture and Technology (main campus) has its own water plant which supplies water for the entire campus.

2.2 Sample Collection

Water samples were collected from various Estates in Nairobi, to test for both THMs and HAAs. The water samples (500mL) were collected into plastic bottles from Kawangware Estate (Nairobi West), Hurlingham, (Nairobi West), Chiromo Campus (University of Nairobi), Catholic University of East Africa (Karen, Nairobi), Kenyatta University (Main Campus), Starehe Boys Centre (Nairobi East), Karura Forest (Kiambu), Kimathi Estate (Nairobi East), Jomo Kenyatta University of Agriculture and Technology (Juja), Kenya Polytechnic University College (Nairobi), hence a total of 10 samples. Samples were collected directly from the tap into the plastic bottles after allowing it to flow for 1 min, filled to the brim, and later refrigerated at 4°C prior to analysis. Analysis was done 6 days after sample collection at Kenya Bureau of Standards (KEBS) at South C, Nairobi Kenya.

2.3 Instrumentation and Experimental Procedures

2.4 Dissolved Organic Carbon

The TOC samples were prepared by placing the sample in 50 mL head-space free vials and acidified below a pH of 2 with phosphoric acid. DOC samples were first filtered through a 0.45 µm filter paper (Cole-Parmer® Nylon Membranes) before acidifying and placing it in the vial. Measurements were performed using a TOC-V CPH analyzer with a Shimadzu ASI-V autosampler and catalytically aided combustion oxidation non-dispersive infrared detector

(NDIR) having a method detection limit of 0.08 mg/L (Shimadzu Corporation, Kyoto, Japan). Operating conditions for DOC measurements were; injector volume 50 μ L; oven temperature 680°C; carrier gas flow 150 mL/min; potassium hydrogen phthalate standards 0-10 mg/L.

2.5 UV Absorbance and Colour

UV absorbance at a wavelength of 254 nm was measured using a HACH DR/4000 UV/VIS spectrophotometer (Hach Company, Loveland, CO). Color was measured using visual platinum method.

2.6 Temperature and pH

Temperature and pH were measured using an Accumet Excel XL50. pH probes were calibrated using standard buffer solutions from Fisher Scientific. The temperature probe was also calibrated using a mercury thermometer.

2.7 Trihalomethane and HAAs Analysis

Briefly, water vials were removed from refrigerated storage and allowed to equilibrate to room temperature before analysis. Immediately after removal of the vial cap, water (10 μ L) was removed using a pre-cleaned gas-tight syringe and transferred into a SPI headspace vial and the SPI vial immediately crimp-sealed using Teflon-lined septum.

Samples were analyzed using SPI/gas chromatography–electron capture detector (SPI/GC-ECD) on a TraceMS (ThermoFisher, Austin, TX) attached to a Trace 2000 gas chromatograph equipped with a split/splitless injector (1177; split) and operated in the splitless mode with a split ratio of 100. Because of the volatility of the THMs, rapid screening of THMs was achieved by direct injection (injection temperature; 250°C) of the water sample into a capillary column (450-type phase, 30m x 0.32mm 1.8 micron, Varian Equipment) connected to ECD. For THMs, this technique produced a short turn-around time with an analysis time of less than eight minutes; furthermore, no sample preparation was required.

The SPI was cooled for injection and then heated rapidly to provide a sharp injection band. The water was unretained, and the initial column temperature was held just above its boiling point to ensure its quick elution. The column oven was then programmed to readily elute the VOCs and

any other higher boiling organics. Injections were made into a pre-column (4m x0.52mm uncoated silica) to retain the salts and prevent column contamination. Automated sampling was done using a Varian auto sampler (8400 Varian Chromatography Systems). The carrier gas was Nitrogen with a constant column flow of 2mL/ min. Delay between injections was one minute and there was cleaning between injections. The sample penetration depth was 90% and for the solvent was also 90%. The default wash volume was 5.0 μ L and the draw up speed was 5.0 μ L/ second. The detector ECD used nitrogen as its make up gas with a flow of 44mL/minute. The set point temperature for the detector was 50 °C, with a cell contact potential of 400mV.

2.8 Determination and quantification of HAAs and THMs

The Xcalibur Quan software (ThermoFisher) was used for peak integration, calibration, and quantification. Peaks were integrated with the integrated collaborative information systems integrator and confirmed by visual inspection. Relative response factors were calculated on the basis of the relative peak areas of analyte quantitation ion and labeled analog ion. Trihalomethanes were quantified by comparing the ratios of analyte peak areas with labeled analog areas for both unknowns and freshly prepared calibrators/standards. The calibration range was 0, 2, 4, 6, 8, 10 mg/L for all the analytes.

3.0 RESULTS and DISCUSSION

Results of the water quality testing are presented in Table 1. A narrow range of turbidity (4.5–4.9 TU) and DOC (1.0–2.3 μ g/L) was recorded. The lowest and highest turbidity values were seen in Kimathi Estate and Hurlingham respectively. The lowest and highest TOC values were seen in Chiromo Campus and Hurlingham respectively.

Table 1 Tap water quality parameters

Water samples	pH	Temp °C	Turbidity (TU)	Colour (TCU)	DOC (μ g/L)	UVA ₂₅₄ (CM ⁻¹)
Chiromo Campus (UoN)	6.9	23.1	4.6	1	1.0	0.162

Catholic University of East Africa (Karen)	7.0	23.6	4.8	2	1.4	0.302
Kawangware Estate (Nairobi West)	7.2	23.9	4.8	3	2.0	0.402
Kenyatta University (Main Campus)	6.9	23.2	4.6	1	1.1	0.161
Starehe Boys Centre (Nairobi East)	6.8	23.0	4.6	1	1.2	0.162
Karura Forest (Kiambu)	7.0	23.5	4.8	2	1.8	0.302
Kimathi Estate (Nairobi East)	6.9	23.1	4.5	1	1.1	0.160
JKUAT (Main Campus)	6.9	23.0	4.7	1	1.1	0.162
Kenya Polytechnic University College	7.1	23.9	4.8	3	2,1	0.403
Hurlingham (Nairobi West)	7.2	24.0	4.9	4	2.3	0.408

Organic matter (NOM) within a given water sample. Colour and UVA_{254} were low an indication that the NOM in the water samples collected had less significant C=C double bond. None of the water samples showed traces for HAAs. HAAs are likely to form at low pH values, from the data obtained in table 1, the pH values were ranging close to neutral and slightly basic. THMs form at basic pHs and its concentration increases with increase in pH, DOC, temperature and chlorine dose in water.

The GC Standards (THMs) in ppm depicted the following peak areas according to the shown retention time (Min)

Table 2.0 Retention Time(s) and Peak Area(s) for Chloroform

#	Name	Time [Min]	Quantity [% Area]	Height [μ V]	Area [μ V.Min]	Area % [%]
1	UNKNOWN	1.37	58.51	62912.7	3491	58.514
2	UNKNOWN	1.57	6.23	2232.9	371.7	6.229
3	UNKNOWN	1.65	3.02	1646.7	179.9	3.015
4	UNKNOWN	1.82	0.47	659.4	27.8	0.467
5	chloroform	1.97	1.8	6832.2	107.4	1.8
6	UNKNOWN	2.26	0.04	4.3	2.2	0.037
7	UNKNOWN	2.53	0.08	134.1	4.8	0.081
8	UNKNOWN	2.6	0.08	333	5	0.084
9	UNKNOWN	2.66	12.04	30921.7	718.1	12.036
10	UNKNOWN	3.14	0.12	40.8	6.9	0.115
11	UNKNOWN	3.88	12.02	23316.5	717.3	12.022
12	UNKNOWN	5.33	0.07	28.8	4.3	0.072
13	UNKNOWN	5.47	4.79	7006.7	285.9	4.792
14	UNKNOWN	6.09	0.15	197.5	8.8	0.148
15	UNKNOWN	8.69	0.28	338.2	16.6	0.278
16	UNKNOWN	9.99	0.31	201	18.5	0.31
Total			100	136806.6	5966.1	100

Table 3.0 Retention Time(s) and Peak Area(s) for dibromochloromethane

#	Name	Time [Min]	Quantity [% Area]	Height [μ V]	Area [μ V.Min]	Area % [%]
1	UNKNOWN	1.2	0.04	135.3	1.8	0.037
2	UNKNOWN	1.38	58.38	63855.3	2829.7	58.375
3	UNKNOWN	1.45	0.8	1150.7	38.8	0.801

4	UNKNOWN	1.55	12.09	1999.9	586.1	12.091
5	chloroform	1.97	4.22	4624.2	204.6	4.22
6	UNKNOWN	2.01	0.34	367.7	16.3	0.337
7	UNKNOWN	2.53	0.12	53.5	5.7	0.119
8	dibromochloromethane	2.66	9.47	19931	458.9	9.467
9	UNKNOWN	2.79	0.25	112	12	0.247
10	UNKNOWN	3.83	0.12	116.4	5.6	0.116
11	UNKNOWN	3.88	9.6	15174.7	465.1	9.595
12	UNKNOWN	4.16	0.06	85.1	3	0.061
13	UNKNOWN	5.33	0.12	58.6	5.6	0.116
14	UNKNOWN	5.47	4.17	4646.2	202.3	4.173
15	UNKNOWN	5.61	0.13	210.2	6.5	0.134
16	UNKNOWN	5.86	0.11	89.6	5.4	0.111
Total			100	112610.4	4847.5	100

Chloroform and BDCM are extremely volatile relative to the other two constituents of THMs and are ultimately transferred to air as a result of their volatility WHO research suggests equal contributions to total chloroforms and BDCM exposure coming from four areas: ingestion of drinking water, inhalation of indoor air largely due to volatilization from drinking water, inhalation and dermal exposure during showering or bathing and ingestion of food.

Table 4.0 Retention Time(s) and Peak Area(s) for bromodichloromethane

#	Name	Time [Min]	Quantity [% Area]	Height [μ V]	Area [μ V.Min]	Area % [%]
1	UNKNOWN	1.2	0.04	135.3	1.8	0.037
2	UNKNOWN	1.38	58.38	63855.3	2829.7	58.375
3	UNKNOWN	1.45	0.8	1150.7	38.8	0.801
4	UNKNOWN	1.55	12.09	1999.9	586.1	12.091

5	chloroform	1.97	4.22	4624.2	204.6	4.22
6	UNKNOWN	2.01	0.34	367.7	16.3	0.337
7	UNKNOWN	2.53	0.12	53.5	5.7	0.119
8	dibromochloromethane	2.66	9.47	19931	458.9	9.467
9	UNKNOWN	2.79	0.25	112	12	0.247
10	UNKNOWN	3.83	0.12	116.4	5.6	0.116
11	bromodichloromethane	3.88	9.6	15174.7	465.1	9.595
12	UNKNOWN	4.16	0.06	85.1	3	0.061
13	UNKNOWN	5.33	0.12	58.6	5.6	0.116
14	UNKNOWN	5.47	4.17	4646.2	202.3	4.173
15	UNKNOWN	5.61	0.13	210.2	6.5	0.134
16	UNKNOWN	5.86	0.11	89.6	5.4	0.111
Total			100	112610.4	4847.5	100

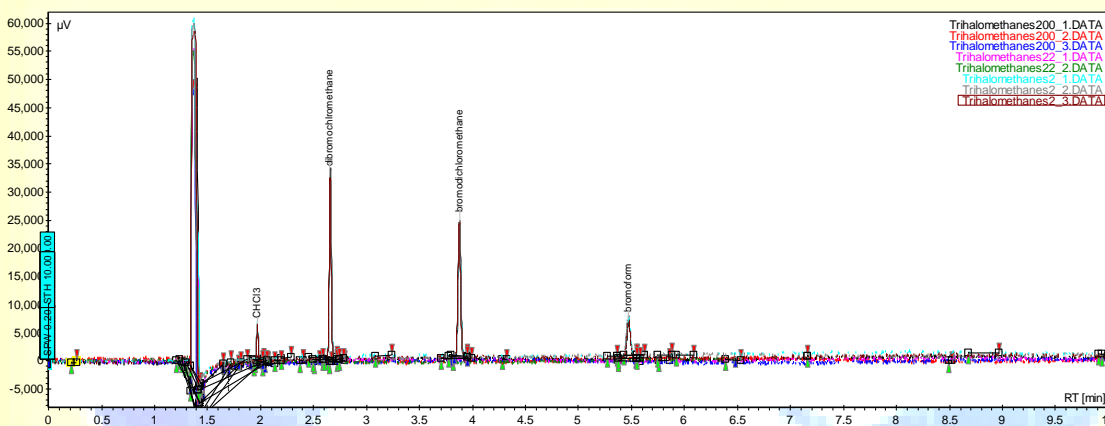
Table 5.0 Retention Time(s) and Peak Area(s) for bromoform

#	Name	Time [Min]	Quantity [% Area]	Height [μ V]	Area [μ V.Min]	Area % [%]
1	UNKNOWN	1.2	0.04	135.3	1.8	0.037
2	UNKNOWN	1.38	58.38	63855.3	2829.7	58.375
3	UNKNOWN	1.45	0.8	1150.7	38.8	0.801
4	UNKNOWN	1.55	12.09	1999.9	586.1	12.091
5	chloroform	1.97	4.22	4624.2	204.6	4.22
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10	UNKNOWN	3.83	0.12	116.4	5.6	0.116
11	bromodichloromethane	3.88	9.6	15174.7	465.1	9.595
12	UNKNOWN	4.16	0.06	85.1	3	0.061
13	UNKNOWN	5.33	0.12	58.6	5.6	0.116
14	bromoform	5.47	4.17	4646.2	202.3	4.173

15	UNKNOWN	5.61	0.13	210.2	6.5	0.134
16	UNKNOWN	5.86	0.11	89.6	5.4	0.111
Total			100	112610.4	4847.5	100

The THMs including CHCl_3 showed a signal at retention time (min) 1.97, CHClBr_2 , CHCl_2Br and CHBr_3 showed their signals at 2.66, 3.88 and 5.47 retention time(s) respectively.

Figure 2 Chromatogram for the GC THMs standards



The Results from all 10 samples analyzed for the four individual THMs are presented in Table 6. Water samples from four locations; Kimathi Estate (Nairobi East), Chiromo Campus (University of Nairobi), Jomo Kenyatta University of Agriculture and Technology (Main Campus) and Starehe Boys Centre, including the blank sample, did not show any traces of THMs. None of the analytes showed traces of bromodichloromethane and bromoform.

Two samples, from Kawangware Estate and Hurlingham both from Nairobi West had some significant amount of Chloroform; $7.4\mu\text{g/L}$ and $13.06\mu\text{g/L}$ respectively. Water samples from Kenya Polytechnic University College (Nairobi Town), Karura Forest (Kiambu), Catholic University of Eastern Africa (Karen, Nairobi), Kawangware Estate and Hurlingham showed traces of dibromochloromethane (BDCM). This included; $6.6\mu\text{g/L}$, $5.6\mu\text{g/L}$, $3.7\mu\text{g/L}$, $6.1\mu\text{g/L}$, $1.7\mu\text{g/L}$ respectively

Table 6 Concentration of chloroform and dibromochloromethane

Sample ID	Chloroform ($\mu\text{g/L}$)	Dibromochloromethane ($\mu\text{g/L}$)
Catholic University of East Africa	-	3.7
Kawangware Estate	7.4	6.1
Karura Forest	-	5.6
Kenya Polytechnic University College	-	6.6
Hurlingham	15.0	1.7

WHO proposes the use of an additive toxicity guideline value (GV), using the fractionation equation that the sum of the four THMs' actual concentration (C) divided by their guideline value (GV) should not be greater than one (WHO 2004).

$$C_{\text{Chloroform}}/GV_{\text{Chloroform}} + C_{\text{BDCM}}/GV_{\text{BDCM}} + C_{\text{DBCM}}/GV_{\text{DBCM}} + C_{\text{Bromoform}}/GV_{\text{Bromoform}} \leq 1$$

Using the results obtained, no sample exceeded the WHO guideline values for any of the four individual THMs. Since the GV value for all the four THMs was 0.0953

Table 7 WHO Guideline values in $\mu\text{g/L}$

Trihalomethanes	WHO Guideline value $\mu\text{g/L}$
Chloroform (CHCl_3)	300
Bromodichloromethane (CHClBr_2)	60
Dibromochloromethane (CHCl_2Br)	100
Bromoform (CHBr_3)	100

CONCLUSION

Water samples from Kawangware Estate and Hurlingham both from Nairobi West had some significant amount of Chloroform; 7.4 $\mu\text{g/L}$ and 13.06 $\mu\text{g/L}$ respectively. Water samples from Kenya Polytechnic University College (Nairobi Town), Karura Forest (Kiambu), Catholic University of Eastern Africa (Karen, Nairobi), Kawangware Estate and Hurlingham showed traces of dibromochloromethane. This included; 6.6 $\mu\text{g/L}$, 5.6 $\mu\text{g/L}$, 3.7 $\mu\text{g/L}$, 6.1 $\mu\text{g/L}$, 1.7 $\mu\text{g/L}$ respectively. The data presented herein clearly show that chlorination of filtered waters does not lead to trihalomethane concentrations that exceed the WHO guideline values as shown in Table 7.

None of the water samples showed traces of HAAs. Proper chlorination of household water does not form harmful levels of disinfection by-products (DBP).

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REFERENCE

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