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Effects of household handling on disinfection by-products

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Abstract

Chlorinated water samples were used to determine the effect of handling modes on disinfection byproducts (DBPs). The DBPs studied were trihalomethanes (THMs), haloacetonitriles (HANs), chloral hydrate (CH), chloropicrin (CP) and 1,1,1-trichloropropanone (TCP). Tap water samples were collected from the distribution system in Damanhour City (Egypt). The investigated strategies included storing water in covered and uncovered bottles in a refrigerator up to 9 hours, with and without previous short boiling. Water quality parameters were not affected by storage or boiling except for electrical conductivity (EC), which decreased after boiling. 90% of THMs were removed by boiling and storage for 9hrs.HANs, including dichloroacetonitrile (DCAN), dibromoacetonitrile (DBAN) and trichloroacetonitrile (TCAN), were not affected by storage, but they were not detected after boiling for 30 seconds. CH and TCP, like the HANs, were affected by boiling rather than storage.

Keywords

Disinfection by products
Drinking water
Water refrigeration
Boiling water
Egypt

1. Introduction

Chlorine is currently the most reliable chemical disinfectant used for water disinfection. However, chlorine also reacts with natural organic matter (NOM) present in water, leading to the formation of trihalomethanes (THMs), haloacetic acids (HAAs), halonitromethanes (HNMs) and other disinfection byproducts (DBPs) **[1]**, **[2]**.Carcinogenic and reproductive effects of DBPs have been reported in toxicological and epidemiological studies **[3]**, **[4]**.THMs are regulated because of their potential health risk, and because they act as surrogates for the control of other halogenated DBPs with health concerns. At the USEPA, the sum of four THMs (THM4, i.e., chloroform, bromodichloromethane, dibromochloromethane, bromoform) is regulated at 80μg/L **[5]**. In the United Kingdom (UK) and Canada, THMs are regulated, with a maximum concentration value of 100μg/L **[6]**, **[7]** which is the same value as in Egyptian regulations **[8]**.After THMs and HAAs, chloral hydrate (trichloroacetaldehyde hydrate) is the next most prevalent disinfection byproduct in drinking water. According to World Health Organization (WHO) guidelines, chloral hydrate should be limited in drinking water because of its adverse health effects and the current MCL is set at 10μg/L **[9]**. The most abundant HANs after water chlorination are dichloroacetonitrile and its brominated analogs, bromochloroacetonitrile and dibromoacetonitrile. Data available on trichloroacetonitrileis insufficient to serve as a basis for defining a guideline value for trichloroacetonitrile. The previous provisional guideline value of 1μg/L was based on developmental toxicity studies. Dichloroacetonitrile induced decreases in body weight and increases in relative liver weight in short-term studies. Despite the potential health effects, there is no US regulatory limit for these compounds, but WHO has suggested guideline values of 20μg/L for DCAN and 70μg/L for DBAN **[9]**.

Indoor handling,such as boiling, refrigerating and storing of drinking water, can greatly impact DBP concentrations.Krasner and Wright **[10]** studied the impact of boiling water on different DBPs. The boiling experiments were conducted on water samples from the Weymouth water treatment plant (California) in the winter of 2000. They found that from 68% to 98% of THMs were removed when chlorinated water was boiled for 1–5 minutes. Complete removal of 1,1,1-trichloropropanone (TCP) was observed after boiling for 1 minute. Chloral hydrate (CH) concentration was reduced by at least 97% following a 1-minute boil of water and was not detected after 2 min. 94–98% removal of the HANs occurred upon boiling the chlorinated samples for 1 minute, and no HANs were detected in the chlorinated water after 2 minutes. 57% removal was observed for CP in water boiled for 1 min, and was not detected after2 minutes of boiling. Battermanet *et al.* **[11]** examined thermal effects on THM concentrations by heating chlorine-free distilled water in an electric kettle. Average removal reported for chloroform and bromodichloromethane at 100˚C were 81% and 73% respectively, while 69% reductions in THM concentration were also reported. Wu *et al.* **[12]** conducted boiling experiments of Seattle tapwater typically containing 0.9mg/L chlorine residual and 0.9–1.5mg/L of dissolved organic carbon (DOC). As boil time increased from 1 to 5minutes, reduction increased from 68% to 83% for chloroform and from 75% to 94% for BDCM. CH and DCAN were not detected after boiling for 1 min. Lahl *et al.* **[13]** reported THM volatilization losses of 73% for a 1minute boiling and 88% following a 5minute boiling. The highest amount removed during boiling reported for chloroform. Kuo *et al.* **[14]** studied THMs and other volatile organic compounds (VOCs) in water samples collected from three cities in Taiwan. They reported THM removal ranged from 61% to 82% upon boiling chlorinated water.

Levesque *et al.* **[15]** studied the effect of storage in refrigeration and boiling of water on THMs. They reported THM reduction due to the storage of water in the refrigerator for 48hr in a covered pitcher, averaging 14%, while reduction by storing water in uncovered pitchers was relatively high, averaging 61%. THM reduction alone by boiling water for 30 seconds averaged 83%. Boiling tap water for 30s and storing it for 48h in the refrigerator in an uncovered pitcher resulted in a 97% average reduction of THMs.

There are a number of studies evaluating different heating, boiling, and filtering devices **[15]**, **[16]**, **[17]**, but there was no available data covering the effect of storage on extended DBPs such as CH, TCP, and HANs. The aim of this study was to investigate the effect of storage and boiling of tap water on some water quality parameters and the concentrations of THMs, CH, HANs and TCP.

2. Materials and Methods

Samples for these experiments were collected from laboratory tap water. Chlorinated water samples were collected after leaving the water running for five minutes. Four different modes for tap water handling were considered: storage of water in the refrigerator in a covered glass bottle (M1.a), storage of water in the refrigerator in an uncovered bottle (M1.b), boiling and storage of water in the refrigerator in a covered bottle (M2.a), boiling and storage of water in the refrigerator in an uncovered bottle (M2.b).

All samples were stored at 4°C. Storage times were estimated at 0hr, 4hr and 9hr. 0hr samples in M1a and M1b were collected directly from the tap water. 0hr samples in M2a and M2b were collected after five minutes when the kettle turned off.

All samples were collected in 1L wide-mouth glass bottles with about 20% air space to match household conditions. For modesM2.a and M2.b, the water was boiled using a plastic kettle. The kettle was turned off 30 seconds after boiling and the water was left to cool in the kettle for five minutes. The results reported represent an average of five experimental dates for studying tap water performed over 5 weeks (**Table 1**).

2.1 Analytical procedures

All measurements were carried out in accordance with the Standard Methods for the Examination of Water and Waste Water **[18]**. All chemicals and reagents were purchased from HACH, Sigma-Aldrish, Chemlab, Merck, Scharlau and Panreac.

Residual chlorine was measured for treated water by photometry(S.M. 4500-Cl G) **[18]** using a HACH colorimeter.Turbidity was measured for water samples using a HACH 2100N turbidity meter (S.M.2130B) **[18]**.

2.2 Analysis of Total Organic Carbon (TOC)

TOC analysis was performed according to (S.M. 5310B) **[18]** for raw and treated water samples. TOC was analyzed with a TOC Analyzer (Tekmar-Dohrmann Apollo 9000). The samples were acidified to a pH less than 2 by phosphoric acid then introduced into the instrument. The samples were purged with high purity hydrocarbon free air for 10 minutes to remove inorganic carbon then injected into a heated reaction chamber packed with a platinum oxide catalyst oxidizer to oxidize organic carbon to CO_2 which was measured by a non-dispersive infrared detector.

2.3 Analysis of DBPs

Trihalomethanes (chloroform, bromodichloromethane, chlorodibromomethane, and bromoform), haloacetonitriles (HANs) (trichloroacetonitrile, dichloroacetonitrile, dibromoacetonitrile), chloropicrin, 1,1,1-trichloropropanone and chloral hydrate were analyzed as described in US-EPA method 551.1 **[19]**.

An Agilent 7890A Gas Chromatograph with an electron capture detector (ECD) with DB-1 30m×0.25mm×1.00µm capillary column was used for identification and quantification of DBPs. The GC injection temperature was 220° C. The ECD temperature was 300° C. The column temperature program was 35° C held for 9 minutes, then a 1° C per minute increase to 40° C which was maintained for 3 minutes, and finally a 6° C per minute increase until a temperature of 150° C was reached, which was held for 1 minute. The injection was splitless with a set time of 0.5 minutes. Flow was set to 23 centimeter/second linear velocity.

3. Results and Discussion

3.1 Effects on water quality parameters

The results for the water quality parameters after boiling and storage are tabulated in **Table 2**. Turbidity increases after boiling (M2.a, M2.b) but decreases with storage time. This result is in agreement with Levesque *et al.* **[15]**. The conductivity results decrease after boiling; this could be due to precipitation of inorganic salts after boiling. A decrease in the concentration of residual chlorine through storage time and after boiling was observed. Residual chlorine decreased from 0.68 mg/L to 0.63 mg/L after 9hrs storage in the covered bottle (M1.a)and to 0.55 mg/L in the uncovered bottle (M2.b). It was not found after boiling.

Table 1: Water characteristics through 5 weeks of baseline water.

Parameter	Unit	Mean	SD	Min	Max
Turbidity	NTU	0.61	0.08	0.51	0.72
Conductivity	µS	432	7.91	422	442
TOC	mg/L	3.5	0.08	3.4	3.6
Free chlorine	mg/L	0.68	0.03	0.64	0.72
CF	µg/L	49.24	3.16	45.3	53.3
DCAN	µg/L	3.2	0.31	2.8	3.6
DCBM	µg/L	16.3	0.32	15.9	16.7
CH	µg/L	7.3	0.32	6.9	7.7
DBCM	µg/L	5.8	0.32	5.4	6.2
THMs	µg/L	71.34	3.8	66.6	76.2
TCP	µg/L	3.7	0.16	3.5	3.9

3.2 Effects on DBPs

The THMs were the most prevalent DBP in the investigated samples followed by CH, while low levels of TCP and DCAN were found. BF and DBAN, CP and TCAN were not detected in all samples.

THM concentrations of the baseline water were reduced by all water handling modes (**Table 3**). THM reduction by the storage of water

Table 2: Average values of water quality parameters for handling modes at storage times.

Handling	Turbidity (NTU)			Conductivity (µS)			TOC (mg/L)			Free chlorine (mg/L)		
modes	0hr	4hrs	9hrs	0hr	4hrs	9hrs	0hr	4hrs	9hrs	0hr	4hrs	9hrs
M1.a	0.61	0.58	0.53	432	436	446	3.5	3.6	3.4	0.68	0.66	0.63
M1.b	0.61	0.59	0.55	432	440	438	3.5	3.3	3.4	0.68	0.61	0.55
M2.a	0.79	0.71	0.58	410	404	409	3.1	3.3	3.1	ND	ND	ND
M2.b	0.79	0.69	0.67	410	462	422	3.1	3.1	3.2	ND	ND	ND

for 9hours in the refrigerator in a covered bottle (M1.a) averaged 12%, while reduction by storing water in an uncovered bottle (M1.b) was relatively high,averaging 30%.The mechanism for reducing THMs consisted mainly of volatilization. These removal percentages were higher than removal reported by Weinberg *et al.* **[20]**. They reported thatrefrigeration of cold tap water in an open container for 12hrs removed only about 8%. Levesque *et al.* **[15]** obtained higher removal of up to 17% for a covered pitcher and 43% for an uncovered pitcher but for 48hr of storage.

Table 3: THMs removal percentagefor all handling modes

Handling modes	THMs removal %		
	0hr	4hrs	9hrs
M1.a	--	4	12
M1.b	--	19	30
M2.a	37	70	84
M2.b	37	88	90

Storage times had a major impact on THM reduction. THM reduction increased as the storage time increased in both covered and uncovered bottles. Average THMresults for 0hr storage were 71.3μg/L. In covered bottles (M1.a), average THMs results decreased from 68.3to 62.4 μg/L after 4hr to 9hr storage, respectively (**Figure 1**). Uncovered bottles (M1.b) show higher THMs reduction, with average THMsresults decreasing from 58to 50.0μg/L after 4hr to 9hr storage, respectively (**Figure 1**).

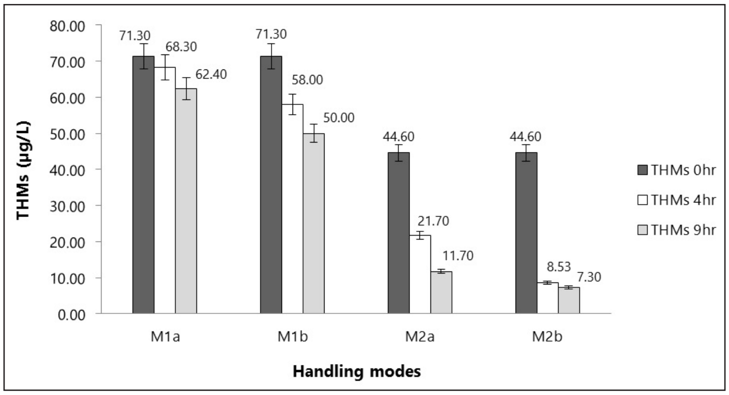

Figure 1: Average THMsresults through storage time in different handling modes.

Although the water contained concentrations of free residual chlorine and organic matter at the time of storage (**Table 2**), there is no further formation of THMs observed. This may be due to the fact that chlorine reactions tend to level off when water temperature decreases.

Boiling water in a kettle for 30 seconds (M2) achieved higher removal percentages of up to 90% of the THM concentration in the baseline water. Reduction of THMs by boilingthe water (M2) without storage removes 37% of THMs (**Table 3**). This result is less than the removal reported by Krasner and Wright **[10]** and Wu *et al.* **[12]** for 1minute of boiling, who reported 74% and 67%, respectively. Levesque *et al.***[15]** reported an 83% reduction after boiling for 30 seconds.

In M2.a, THMs removal increases from 70 to 84% after 4hrs and 9hrs, respectively. A slightly higher THMs removal was observed in M2.b from 88 to 90% after 4hrs and 9hrs, respectively (**Table 3**). Boiling water seems to be the fastest procedure to remove THMs without storage.

All other detected DBPs such as CH, DCAN and TCP show the same behavior toward the handling modes. Since these DBPs are not volatile compounds, storage of water (modesM1.a and M1.b) did not have any effect on their concentrations. Average concentrations remained practically the same during the time of storage (4 or 9hrs), compared to those observed in the baseline water concentration (**Table 4**).

After boiling in a kettle for 30 seconds (M2.a and M2.b), all concentrations of CH, DCAN and TCP were below the limit of quantification (0.5μg/L) (**Table 4**). This shows the significant effect of boiling on these DBPs. These results are consistent with other research which has studied the effect of boiling on DBPs **[10]**,**[12]**, **[15]**, **[20]**.

Table 4: Average concentrations of CH, TCP and DCAN in different processes with storage time.

	TCP(μg/L)			CH(μg/L)			DCAN(μg/L)		
	0hr	4hrs	9hrs	0hr	4hrs	9hrs	0hr	4hrs	9hrs
M1.a	7.3	7.5	7.2	3.2	2.9	2.9	3.7	3.5	3.5
M1.b	7.3	7.2	7.2	3.2	3.1	2.9	3.7	3.4	3.3
M2.a	ND	ND	ND	N.D	N.D	N.D	N.D	N.D	N.D
M2.b	ND	ND	ND	N.D	N.D	N.D	N.D	N.D	N.D

4. Conclusion

Carcinogenic and reproductive effects of DBPs have been reported in toxicological and epidemiological studies. The indoor handling modes investigated in this study showed great impact on disinfection byproducts. Boiling had the greatest removal ratio for all compounds. THMs were significantly removed by boiling due to their volatility. CH, TCP and DCAN were totally removed after boiling. Storage and refrigeration achieved lower removal only with THMs. CH, TCP and DCAN were not affected by storage and refrigeration.Despite the simplicity of these handling modes, it shows important implications in reducing exposure to the adverse health effects of disinfection byproducts.These results concern only a small sample and further study is necessary for other byproducts such as haloacetic acids.

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