

Effects of tap water processing on the concentration of disinfection by-products

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ABSTRACT

Aim: This study examined the effects on disinfection by-product (DBP) concentrations of common household methods for processing drinking water.

Methods: We investigated the effects of refrigerator storage, jug filtering, boiling in an electric kettle, and supply from an instant boiling water unit, with or without filtering, on four species of trihalomethanes (THMs) and nine species of haloacetic acids (HAAs) in water ready for consumption in Sydney, Australia. Water samples were processed in such a way as to simulate real life conditions for drinking filtered water or hot water drinks prepared from tap water drawn from public water supply systems.

Results: There was a large reduction in total THMs in kettle-boiled water, instant boiled water, jug-filtered water and instant boiled-filtered water (reductions of 85.8, 93.5, 92.6 and 87.8% of their concentration in tap water respectively). Refrigerator storage did not appear to have a consequential effect on THMs or HAAs. Jug-filtering and instant boiling and filtering resulted in large decreases (77–94%) in all species of HAAs in tap water.

Conclusion: This study suggests that different methods of processing tap water can change DBP concentration to an extent that would have a meaningful impact on exposure assessment in epidemiological studies.

Key words | boiling, disinfection by-products, filtering and refrigerator storage, water handling

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INTRODUCTION

Disinfection by-products (DBPs) in drinking water are formed as a result of chemical reactions between the disinfectant (chlorine, bromine or ozone) and naturally occurring organic matter in the water. There is some evidence, although inconclusive, for associations between human exposure to chlorination DBPs and adverse health effects, including cancers of the bladder (Villanueva *et al.* 2004), colon and rectum (Morris *et al.* 1992; Morris 1995; King *et al.* 2000; Bove *et al.* 2007; Rahman *et al.* 2010) and kidney (Morris *et al.* 1992), and reproductive and developmental effects (Nieuwenhuijsen *et al.* 2000).

In most studies of health effects of DBPs, an ecological measure of exposure, such as DBP concentration in a water

supply distribution system, has been used as a surrogate measure of individual exposure. However, DBP levels can vary in a distribution system depending on temperature, distance from the point of chlorination, and the amount of organic matter in the water (Villanueva *et al.* 2007). Thus individuals' exposure will vary depending on the concentration of DBPs in the water they drink at home, work and elsewhere. It will also vary depending on how water is processed shortly before it is consumed; the nature and concentration of DBPs in drinking water may change when tap water is boiled, filtered with a point of use device (e.g. filter jugs, plumbed kitchen filters) or stored in a refrigerator (Wu *et al.* 2001; Krasner & Wright 2005; Levesque *et al.* 2006; Weinberg *et al.* 2006).

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Some studies have demonstrated a decrease (68–98%) in trihalomethanes (THMs) after boiling water for periods of 0.5–5 min (Wu *et al.* 2001; Batterman *et al.* 2000; Krasner & Wright 2005; Levesque *et al.* 2006) and removal of large amounts of THMs (47–99%) by filtering the water through activated carbon or ion exchange resin filters (Gibbons & Laha 1999; Levesque *et al.* 2006; Weinberg *et al.* 2006). Storing chlorinated tap water in a refrigerator for four hours in a covered or uncovered plastic pitcher resulted in 15 and 27% reductions in THMs, respectively (Levesque *et al.* 2006). A study by Krasner & Wright (2005) found a 19% increase in total haloacetic acids (THAAs) in chlorinated water but a 22% decrease in chloraminated water after boiling for one minute. Wu and co-workers (2001) found a two-fold increase in dichloroacetic acid (DCAA) but a 30% decrease in trichloroacetic acid (TCAA) in chlorinated water after boiling for one minute. Levesque *et al.* (2006), however, found no material change in HAA concentration after 0.5 min of boiling. Like THMs, HAAs also showed large reductions (68–95%) after filtering (Gibbons & Laha 1999; Levesque *et al.* 2006; Weinberg *et al.* 2006). Refrigerator storage appeared to have no appreciable effect on HAA concentrations (Levesque *et al.* 2006; Weinberg *et al.* 2006).

The generalisation of these study results is limited in a number of respects. Several studies had or appeared to have very small sample sizes (some not reported) (Gibbons & Laha 1999; Krasner & Wright 2005; Levesque *et al.* 2006; Weinberg *et al.* 2006). Two studies only considered THMs (Kuo *et al.* 1997; Batterman *et al.* 2000; and one study used artificial samples containing DBP concentrations much higher than those currently measured in most drinking water supplies (Wu *et al.* 2001). Moreover, the use of different summary statistics and considerable inconsistency in the proportional change reported in these studies prevent quantitative summation of the effects of different processing methods on DBP levels, either in total or for individual chemical species.

Most of the relevant studies also measured the effect of boiling water for one to five minutes. In Australia and many other high income countries, water for hot drinks is boiled in an electric kettle or an instant boiling water unit (ZIP Industries 2009). Most electric kettles manufactured for domestic use keep water at a rolling boil for less than one minute (typically about 15 s) while the instant boiling

water units can keep water at ~100 °C for long periods (minutes to hours), depending on how frequently they are used. Some instant boiling water units have a built-in filtration unit. There is no published literature on the effects of these newer devices on drinking water DBP levels.

Estimates of individual intake of DPBs are commonly based on information on DBP concentrations in tap water, self-report of water consumption and information regarding how the water was processed (treatment plant) before it was consumed. However, even when this information is available, it is insufficient to accurately quantify individual intake of DBPs from drinking water. We undertook to determine the effects on drinking water DBP concentrations of different approaches to water processing that are now in common use in households or workplaces.

METHODS

Study design and sample size calculations

We conducted a cross-sectional survey of chloraminated water drawn from drinking water taps in 18 workplaces and two residences in Sydney, Australia. Four water treatment approaches were tested: water stored in a closed vessel in a refrigerator for 5 h; water filtered using a domestic jug filter; water boiled in a domestic electric kettle for 15 s; and water obtained from an instant boiling water unit, with or without a built-in filter. Where possible, water samples in each location were tested using each approach.

We separately estimated the sample sizes required to detect expected differences in mean DBP concentrations in tap water and processed water for each processing method. Estimations were based on having 80% power to detect a difference at a 5% significance level, using a paired t-test. Resource limitations dictated that we use the minimum sample sizes necessary for the study. Values for the standard deviations and mean differences expected were obtained from a pilot study, which we conducted before the field survey, and from existing literature (Levesque *et al.* 2006). The expected mean difference for refrigerator storage was very low (3%). Thus for this process we based sample size calculations on detecting a 50% change in concentration. For other processes, the expected differences were much greater

(from a 90% reduction of THMs in filtered water to a 40% increase in DCAAs in instant boiling water), and for these we aimed for a sufficient sample to detect the expected mean difference. While samples were collected from 20 locations, fewer samples were analysed for some DBP species and processing combinations when they were sufficient to meet our power requirements as indicated by the numbers given in [Table 1](#).

Collection and processing of water samples

Tap water samples

The tap water samples were collected according to a pre-specified protocol to avoid air bubbles and air spaces in the samples. To stabilise the sample and to avoid further formation of DBPs, 1ml of a 20% sodium thiosulphate solution was added to each sample. Separate samples were collected for analysis of HAAs and THMs. Samples were stored on ice for transportation and in a refrigerator (4 °C temperature or less) pending transport to the laboratory.

Refrigerator storage samples

Samples for refrigerator storage were collected directly into large closed containers with about 5% air space, placed in a refrigerator and held at a temperature of 4 °C for five hours. Samples for analysis were then collected from the containers and treated as above.

Jug-filtered samples

Tap water was collected directly into a domestic jug fitted with ion exchange and activated carbon filtration. Such filter systems are designed to reduce heavy metals, carbonate hardness, tastes and odours (such as those of chlorine), some pesticides and organic impurities. Samples were collected for analysis immediately after approximately one litre of water had passed through the filter.

Kettle-boiled water

Before sample collection, we tested a domestic electric kettle and noted that it kept water at a rolling boil for 10–15 s

before automatically switching off (three tests). One litre of tap water was boiled in the kettle, poured into a container and kept at room temperature for 5 min to reflect real-life conditions for drinking hot beverages. Samples for analysis were then collected, sealed, kept at room temperature until they were just warm to touch and then placed on ice or refrigerated.

Instant boiled and instant boiled and filtered water

One litre of water was collected from each instant boiled or instant boiled and filtered water unit into a container and kept at room temperature for 5 min before samples were collected for analysis as described above for kettle-boiled water. These units are in common use in Australia and many other countries ([ZIP Industries 2009](#)).

All the samples were processed in situ, and all the filters and kettles were the same. The instant boilers were not the same model but were functionally the same.

Analysis of water samples

The water samples were analyzed in a laboratory with Australian National Association of Testing Authorities accreditation for the analysis of DBPs in drinking water. The laboratory also carries out routine DBP analyses for the Sydney water supply utility using the United States Environmental Protection Agency methods for analysis ([EPA 2008a, b](#)). Nine species of haloacetic acids (bromoacetic acids (BAA), bromochloroacetic acids (BCAA), bromodichloroacetic acids, chloroacetic acids (CAA), chlorodibromoacetic acids (CDBAA), dibromoacetic acids (DBAA), DCAAs, tribromoacetic acids and TCAA) and four species of trihalomethanes (chloroform, bromodichloromethane (BDCM), dibromochloromethane (DBCM) and bromoform) were analyzed. The degree of measurement error in the laboratory was reported as $\pm 30\%$ for HAAs and $\pm 25\%$ for THMs.

Statistical analysis

We calculated geometric mean DBP concentrations for the water pre and post household treatment and also calculated the applicable percentage reductions or increases in DBPs.

Table 1 | Mean concentrations of water disinfection by-products in water drawn straight from the water supply (tap water) and drinking water after household or workplace processing

Disinfection by-product species ($\mu\text{g/L}$)	Type of water processing – geometric mean (95% CI)											
	Tap water ($n = 15 \& 14$) ^a		Refrigerator stored water ($n = 7 \& 9$) ^a		Kettle-boiled water ($n = 15 \& 5$) ^a		Instant boiled water ($n = 11 \& 5$) ^a		Jug-filtered water ($n = 8 \& 5$) ^a		Instant boiled-filtered water ($n = 4 \& 5$) ^a	
	Mean (95% CI)	Mean (95% CI)	<i>p</i> -value ^b	Mean (95% CI)	<i>p</i> -value ^b	Mean (95% CI)	<i>p</i> -value ^b	Mean (95% CI)	<i>p</i> -value ^b	Mean (95% CI)	<i>p</i> -value ^b	
Dichloroacetic acids (DCAA)	10.8 (10.1, 11.6)	10.6 (10.1, 11.1)	0.141	12.0 (7.1, 20.6) ^c	0.612	12.4 (9.9, 15.4)	0.134	1.9 (1.0, 3.8) ^c	0.001	2.6 (0.3, 20.6) ^c	0.119	
Trichloroacetic acids (TCAA)	6.9 (6.3, 7.0)	6.6 (6.1, 7.1)	0.468	5.0 (4.6, 5.3)	<0.001	0.4 (0.2, 0.8) ^c	<0.001	1.2 (0.8, 2.1) ^c	0.001	0.4 (0.1, 1.4) ^d	0.009	
Bromochloroacetic acids (BCAA)	3.5 (3.2, 3.8)	3.3 (2.9, 3.7)	0.095	3.7 (3.4, 4.0)	0.221	1.6 (0.9, 2.8) ^c	0.019	0.4 (0.2, 0.8) ^d	<0.001	0.5 (0.1, 1.8) ^c	0.018	
Total haloacetic acids (THAA) ^e	21.0 (19.7, 22.2)	20.4 (19.1, 21.7)	0.163	22.6 (19.2, 26.6)	0.346	14.8 (11.8, 18.6)	0.009	3.9 (2.6, 5.8)	<0.001	4.2 (1.3, 13.9)	0.027	
Chloroform	33.4 (29.8, 33.5)	32.2 (26.8, 37.3)	0.414	4.1 (3.2, 5.4)	<0.001	1.9 (0.4, 8.6)	0.007	2.3 (1.6, 3.4)	<0.001	3.7 (0.6, 25.1) ^c	0.035	
Bromodichloromethane (BDCM)	15.8 (15.3, 16.4)	14.9 (12.4, 17.8)	0.506	2.8 (2.2, 3.5)	0.004	0.5 (0.1, 2.0) ^c	0.005	0.9 (0.4, 2.1) ^c	0.002	1.0 (0.2, 6.3) ^c	0.016	
Dibromochloromethane (DBCM)	4.8 (4.6, 5.1)	4.8 (4.2, 5.5)	0.828	0.4 (0.1, 1.1) ^d	0.005	0.4 (0.1, 1.1) ^d	0.003	0.4 (0.1, 1.1) ^d	0.005	0.4 (0.2, 1.3) ^c	0.006	
Total trihalomethanes (TTHM) ^e	52.3 (50.2, 54.5)	51.3 (43.5, 60.5)	0.468	7.4 (5.9, 9.3)	<0.001	3.3 (1.0, 10.7)	0.003	3.8 (2.6, 5.7)	<0.001	6.2 (1.5, 25.7)	0.014	

^aNumbers of samples for haloacetic acids and for trihalomethanes, respectively, as per power calculations.

^b*p*-values from the paired t-test of the means of tap water and processed water.

^cSamples below the detection limit were randomly assigned a value from a uniform distribution of values from 0 to 1 (detection limit 1 $\mu\text{g/L}$) for the following samples:

- Kettle-boiled water – DCAA (1);
- Instant boiling water – TCAA (9), BCAA (2), BDCM (4);
- Jug-filtered water – DCAA (1), TCAA (1), BDCM (1);
- Instant boiling filtered water – DCAA (1), BCAA (3), chloroform (1), BDCM (3), DBCM (4).

^dNot detectable in all or all but one sample.

^eTHAA is the sum of dichloroacetic acids, trichloroacetic acids and BCAA; TTHM is the sum of chloroform, BDCM and DBCM.

The statistical significance of differences between DBP concentrations in the tap water before and after processing was analysed using paired *t*-tests. Geometric means were used because the data were not normally distributed. Values below the detection limit (detection limit was 1 µg/L), which were common for some species (e.g. 9 out of 11 in TCAA in instant boiling water), were substituted using a value from a distribution, assuming that all the non-detectable values were uniformly distributed between zero and the detection limit (Lin & Niu 1998). In some cases (e.g. DBCM in kettle-boiled water) all or all but one values were below the detection limit and the paired *t*-test was done on the simulated values. The final means and 95% confidence intervals (CIs) were obtained by running 10,000 simulations for each measurement with a value below the detection limit (see Appendix 1, available online at <http://www.iwaponline.com/jwh/009/155.pdf>).

RESULTS

Results are presented as absolute values (geometric mean DBP concentrations) (Table 1), and as percentages (of

the concentration in water drawn straight from the water supply) remaining after processing (Table 2), with their 95% CIs. Among the nine species of haloacetic acids and four species of THMs analysed, six HAAs (CAA, BAA, CDBAA, bromo DCAAs, DBAA and tribromo acetic acids) and one species of trihalomethane (bromoform) had values below the detection limit for all tap water samples and all processed samples. Therefore, no analyses were possible for these species. Of the remaining species, concentrations for some observations were below the detection limit of 1 µg/L. We obtained concentrations for total THMs and total HAAs by summing the detectable subspecies (three subspecies of HAAs and three of THMs). It should be noted that there were different numbers of samples for some of the treatment processes and for different DBP species for reasons stated in the Methods.

Overall, the concentrations of DBPs in tap water varied little from one sample to another, as shown by the generally narrow 95% CIs (Table 1). The mean concentrations of THMs were consistently and substantially reduced by jug-filtering and boiling. Only filtering consistently reduced the concentration of HAAs (Tables 1 and 2); kettle boiling did

Table 2 | Percent disinfection by-product concentrations in water after processing in different ways relative to corresponding concentrations in unprocessed tap water samples (based on values reported in Table 1)

Disinfection by-product species (µg/L)	Refrigerator-stored water % (95% CI) ^a (n = 7 & 9) ^b	Kettle-boiled water % (95% CI) ^a (n = 15 & 5) ^b	Instant boiling water % (95% CI) ^a (n = 11 & 5) ^b	Jug-filtered water % (95% CI) ^a (n = 8 & 5) ^b	Instant boiling filtered water % (95% CI) ^a (n = 4 & 5) ^b
Dichloroacetic acids (DCAA)	90.7 (78.7, 104.5)	111.6 (65.2, 191.1)	117.1 (94.4, 145.4)	16.8 (8.3, 34)	22.5 (2.4, 211.6)
Trichloroacetic acids (TCAA)	96 (84.3, 109.3)	74.7 (67.8, 82.2)	6.5 (3.4, 12.3)	18.7 (10.6, 33)	5.7 (1.5, 22.0) ^c
Bromochloroacetic acids (BCAA)	85.6 (70.7, 103.7)	106.4 (95.9, 118.0)	47.0 (25.9, 85.3)	9.9 (4.5, 21.6) ^c	11.7 (3.0, 45.4)
Total haloacetic acids (THAA)	91.3 (79.4, 105.0)	107.7 (91.6, 126.6)	71.9 (57.1, 90.4)	17.8 (11.1, 28.6)	19.3 (5.4, 69.4)
Chloroform	94.7 (81.8, 109.6)	7.9 (2.8, 22.4)	6.4 (1.5, 27.9)	7.2 (4.9, 10.7)	12.4 (2.0, 77.7)
Bromodichloromethane (BDCM)	95 (80.2, 112.6)	18.0 (14.3, 22.6)	7.7 (2.6, 22.7)	6.1 (2.7, 14.0)	6.1 (0.9, 41.1)
Dibromochloromethane (DBCM)	98.8 (87.2, 111.9)	7.9 (2.7, 23.3)	3.3 (0.8, 13.0)	7.8 (2.6, 23.0) ^c	9.3 (3.2, 27)
Total trihalomethanes (TTHM)	95.2 (82.0, 110.5)	14.2 (12.0, 16.9)	6.5 (2.1, 20.8)	7.4 (4.9, 11.0)	12.2 (3.0, 49.1)

^aPercentage of the geometric mean concentration in source tap water (based on values in Table 1).

^bNumbers of samples for haloacetic acids and for trihalomethanes respectively as per power calculations.

^cNot detectable in all or all but one sample.

not reduce total HAAs but instant boiling did. Boiling appeared to reduce mean TCAA concentrations but increased mean dichloroacetic acid concentrations. BCAA concentrations were substantially reduced in instant boiled water but not in kettle-boiled water. The changes in THAA concentrations in instant boiled-filtered water were similar to those seen with jug filtration. In contrast, for THMs, there was a smaller reduction in instant boiled-filtered water than in jug-filtered water; but the small number of samples makes this apparent difference between HAAs and THMs difficult to interpret. There was little or no change in geometric mean DBP concentrations with closed storage in a refrigerator.

The geometric mean DPB concentrations in tap water expressed as a percentage of their mean concentrations in Table 2 are the processing weights that could be used when estimating individuals' intakes of DBPs.

DISCUSSION

Large reductions in concentrations of all species of DBPs were observed after filtering water and using water from instant boiler units that had a built-in filtration unit. For filtered water, the reduction in concentration was substantial and statistically significant for all DBP species. For kettle-boiled and instant boiled water, the reduction in concentration was large for THMs, but there were small increases for some HAAs. Refrigerator storage did not substantially reduce any of the DBP species.

The fact that filtration reduced THM concentrations more than HAA concentrations was also observed by Levesque *et al.* (2006), and the amount of reduction we observed was very similar to previous studies (Levesque *et al.* 2006; Weinberg *et al.* 2006). The reduction in THMs due to boiling water was comparable to that reported by Krasner & Wright (2005) (although the authors may well have meant heating rather than a rolling boil), and was a little greater than that reported by Levesque *et al.* (2006). The decrease in THMs reflects their volatility (Batterman *et al.* 2000; Wu *et al.* 2001). Although THM formation occurs during the first minute of boiling, the volatilization becomes dominant after this, which results in an overall reduction in concentration (Wu *et al.* 2001).

The increase in DCAA and BCAA in kettle-boiled water and DCAA in instant boiled water is consistent with a previous study where chloraminated tap water was boiled in a kettle (Krasner & Wright 2005). This study found a 9% decrease in DCAA and 4% increase in BCAA after one minute boiling. However, in contrast to that study, we found a 53% reduction of BCAA in instant boiled water; all of our samples came from chloraminated tap water. Therefore, it appears that kettle boiling and the production of instant boiled water might have very different effects on BCAA. It is possible this is because a longer time at a high temperature is required for the breakdown of this HAA sub-species. Interestingly, in Krasner & Wright's (2005) study, boiling reduced total HAAs in chloraminated water but resulted in an increase in chlorinated water. Levesque *et al.* (2006) also found a 35% increase ($p < 0.001$) in DCAA levels after boiling chlorinated tap water but found a 42% decrease ($p < 0.001$) in TCAAs. This was thought to be due to the chlorine residual reacting with the DBP precursors during boiling within the first minute to form additional DCAAs, which are not degraded further (Krasner & Wright 2005; Levesque *et al.* 2006), and decarboxylation of TCAA by chloroform to favour a decrease in TCAA levels (Zhang & Minear 2002).

Processing water by storage in a refrigerator resulted in little or no change in DBP concentrations, although all the levels were decreased. The latter observation is consistent with the study by Levesque *et al.* (2006) which demonstrated an approximately 3% decrease of HAA after 4 h of storage. For THMs, however, Levesque *et al.* (2006) observed a reductions of 15 and 27% respectively following storage of chlorinated water in covered or uncovered plastic pitchers. This inconsistency with our findings could be due to the fact that our water was chloraminated.

Our study has several limitations. The limited sample sizes for refrigerator storage meant the study was under-powered to allow precise assessment of the observed 9 and 5% falls in arithmetic mean total HAAs and total THMs. The relatively high detection limit of the analytical laboratory resulted in many observations being reported as non-detectable, which prevented us from precisely estimating the effects of filtering and boiling, methods which were associated with considerable decreases in DBP concentrations.

That we took sufficient samples to provide adequate power to detect expected changes in DBP concentrations for most categories of water processing is a strength of this study. However, many of the 95% CIs around the estimates of percentage change were wide and considerably larger sample sizes would have been required to estimate the changes precisely. The statistical method used to deal with the non-detectable values (substitution from a distribution and simulation) allowed us to retain some samples with missing values, but at the cost of increasing measurement error.

This study has generated several new findings. They include the evidence of a small increase in the concentration of DCAA in boiled chloraminated water; and clear differential effects of kettle boiling and use of an instant boiling unit on concentrations of TCAAs and BCAAs. Studies with larger sample sizes that also measure DBP precursors and environmental factors affecting DBPs will provide more precise estimates of the effects of processing water.

Our findings have important implications for individual-level exposure assessments in epidemiological studies that combine the concentration of DBPs in tap water with daily water use behaviour to estimate DBP uptake. We suggest that when estimating total DBP intake account is taken of the effects of household or workplace processing on DBP concentrations along with water consumption information and measurement of DBP concentrations in tap water. We have used these results to examine the extent of misclassification of exposure to DBPs when the effect of household and workplace water processing is not considered in a related investigation in 114 Australian subjects with exposure assessment covering all sources (including home and work) and both questionnaire recall and diary recording (unpublished data). Adjusting for water processing has the potential to materially increase the accuracy of estimates of exposure in epidemiological studies of the health effects of DBP exposure.

CONCLUSION

Different methods used for processing household drinking water after it is drawn from the supply, particularly boiling and filtering, can materially alter DBP concentrations.

These changes differ depending on the type of DBP (HAAs or THMs) and, for HAAs, the species, and the type of treatment used for the water supply as a whole (chlorinated or chloraminated). Collection of data on water processing and its use to adjust estimates of individual DBP uptake should be considered in epidemiological studies of the health effects of exposure to DBPs.

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