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Abstract

Background: The chlorination of water has led to a substantial reduction in waterborne disease outbreaks. There are however concerns regarding the safety of trihalomethanes (major by-products of chlorination). One of the limitations of much of the existing evidence is the lack of individual trihalomethane exposure estimates, and these estimates should include the concentration of trihalomethanes in bottled water.

In Australia, water advertised as 'spring' or 'mineral' water should, by definition, remain untreated, and should not, therefore, contain any trihalomethanes. We tested this assumption by assessing the concentration of trihalomethanes (bromodichloromethane, bromoform, chloroform, dibromochloromethane, and total trihalomethanes) in the six brands of bottled spring water most commonly consumed by pregnant women in Australia (here labelled A-F).

Methods: For each water brand, two bottles were purchased from five locations, and this procedure was replicated approximately two weeks later. Standard water analysis techniques were used to determine the concentrations of bromodichloromethane, bromoform, chloroform, dibromochloromethane, and total trihalomethanes in each bottle.

Results: All 10 samples of brands A and B were positive for trihalomethanes, as was one bottle of brand C. No trihalomethanes were detected in any of the samples of the remaining three brands (D-F). The highest recorded total trihalomethane concentration was 30µg/L.

Discussion: The trihalomethane concentration in Australian bottled spring water cannot be assumed to be zero. Studies estimating trihalomethane exposures should therefore collect data on the brand of bottled water consumed, and test these brands contemporaneously for their trihalomethane exposures, to strengthen the evidence regarding trihalomethane exposure and human health outcomes.

Key works: disinfectant by-products, drinking water, dosage, exposure, trihalomethanes, water quality

1. Background

Chlorination of water has led to a substantial reduction in waterborne diseases (Wigle 1998), but there is evidence to suggest an association between a major class of disinfectant by-products, trihalomethanes (THMs), and a range of human health outcomes, including bladder, colon and rectal cancer (Benmarhnia et al. 2018). Maternal THM exposure has been associated with adverse birth outcomes, such as pregnancy loss (Mashau et al. 2018), stillbirth (Rivera-Nunez et al. 2018), pre-term birth (Rivera-Nunez et al. 2018), intrauterine growth retardation (Mashau et al. 2018), compression of the umbilical cord (Rivera-Nunez et al. 2018), placental separation and haemorrhage (Rivera-Nunez et al. 2018), fetal hypoxia/asphyxia (Rivera-Nunez et al. 2018), and congenital anomalies (Nieuwenhuijsen et al. 2009). Establishing the association between THM exposure and human health outcomes is critical, particularly if we are to manage the increased risks associated with climate change: because THM formation increases with temperature, it has been calculated that the 1.8°C temperature rise expected by 2050 will result in a 39% increase in THM formation in public water supplies (Valdivia-Garcia et al. 2019).

The quality of exposure data is often a limiting factor in environmental epidemiology, and the evidence for adverse health outcomes from THM exposure is no exception (Legay et al. 2010; Nieuwenhuijsen et al. 2009). Pregnant women may be exposed to THMs through ingestion, inhalation and dermal absorption (Nieuwenhuijsen et al. 2000; Reif et al. 1996; Villanueva et al. 2006; Weisel and Jo 1996), and the individual exposure to THMs via these routes should be considered, as should the type of water consumed. Total THM exposure estimates should include the type of water (e.g. tap, rain, bore, bottled, boiled tap water) and the route of exposure (e.g. drinking, bathing, showering, swimming, washing dishes). Although a number of previous studies have assumed that bottled water does not contain any THMs (Iszatt et al. 2011; Patelarou et al. 2011; Villanueva et al. 2007; Wright et al. 2006), this assumption should be determined locally, with some studies detecting THMs in bottled water internationally (Ahmad and Bajahlan 2009; Al-Mudhaf et al. 2009; Ayoub Momani 2006; Fakour and Lo 2019; Font-Ribera et al. 2010; Genisoglu et al. 2019), in some cases (Ahmad and Bajahlan 2009; Al-Mudhaf et al. 2009; Ayoub Momani 2006; Fakour and Lo 2019) exceeding 10 µg/L (the limit indicated in International Bottled Water Association's code of practice (International Bottled Water Association 2015)). In Australia the terms 'spring water' and 'mineral water' are reserved from groundwater in its natural state (Australian Government 2017), and should not therefore contain any THMs. Trihalomethane concentrations have not previously been investigated for bottled water sold in Australia, with bottled water potentially including tap water, filtered/purified water or spring water.

As part of a study into maternal exposure to THMs and birth outcomes in Australia, we sought to ascertain the THM exposure related to drinking bottled water. We therefore assessed the concentration of bromodichloromethane, bromoform, chloroform, dibromochloromethane, and total THMs in the six most commonly consumed types of bottled water by pregnant women in Australia.

2. Methods

As part of a larger study into the association between maternal exposure to water disinfectants and birth outcomes (ethics approval from the University of Western Australia Human Research Ethics Committed RA/4/1/4973 and RA/4/1/5280), women in their first trimester of pregnancy were asked to report on their consumption of bottled water, including the three most common brands of bottled water consumed at work and at home. The six most commonly consumed brands at home and at work, based on the 452 women who responded to the questionnaire from June 2012 to June 2014, were brands here labelled A to F for anonymity. All brands sampled were 'spring' water.

In December 2014, two bottles of each of these six brands of water were purchased from six locations in Adelaide, Australia on two days, approximately two weeks apart. The bottles were selected from the front of the shelf, with no effort made to select different expiry dates or batch numbers. On the day of purchase the bottles were taken to SA Water's Australian Water Quality Centre for analysis. The original bottles were used rather than sampling bottles to avoid blow-off of THMs upon opening.

The concentrations of bromodichloromethane, bromoform, chloroform, dibromochloromethane, and total THMs were determined using standard procedures (as per the National Association of Testing Authorities, Australia procedures ISO/IEC 17025 (National Association of Testing Authorities Australia)).

3. Results

No THMs were detected (at a detection limit of <1µg/L for the individual types of THMs, and <4 µg/L for total THMs) in any of the bottles for three (D-F) of the six brands tested. Trihalomethanes were detected in one bottle of brand C, six bottles of brand B, and all 10 bottles of brand A (Table 1). The results for bottled water from brand A were reasonably consistent with a range of 23-28 µg/L detected for the total THM concentration. The results for brand B were less consistent with a range of 0-30 µg/L detected for total THM concentration, as well as ranges exceeding five µg/L for the bromodichloromethane and dibromochloromethane concentrations. There were three distinct concentration patterns for brand B that matched three distinct types of batch numbers (Table 2). Four of the bottles in brand B had batch numbers resembling those of brand A, all of which had similar THM concentrations (Table 2).

4. Discussion

We examined the THM concentrations of the six commonly consumed brands of bottled water among pregnant women. All of the six brands investigated are marketed as 'spring' or 'mineral' water, and should therefore not have contained any THMs as Australian legislation restricts this term to groundwater in its natural state (Australian Government 2017). Despite this, brands A and B consistently contained THMs, with one bottle of brand C also having chloroform detected. It is therefore not possible to assume a concentration of zero when estimating exposure to THMs for such water products in Australia.

The International Bottled Water Association's (2015) code of practice limits total THM concentrations to 10 µg/L. However, our results indicate that this concentration of total THMs cannot be assumed in bottled water sole in Australia, with 14 bottles exceeding this industry limit. The highest total THM concentration reported as 30 µg/L, with 7 µg/L the highest concentration of bromodichloromethane, 14 µg/L for bromoform, 2 µg/L for chloroform, and 7 µg/L for dibromochloromethane. It must be emphasised that these concentrations are all well within the limits stipulated by Australia's National Health and Medical Research Council (2017), as well as the World Health Organization (2017) (see Table 3 for acceptable ranges). The concentrations were also within stipulated guidelines internationally (Jackson et al. 2008), with the exception of the Maximum Contaminant Level Goal (United States) for bromodichloromethane and bromoform (0 µg/L), and total THMs in the Netherlands (25 µg/L; (see Table 3 for the reported concentrations).

The results of this study indicate that the water in brands A and B is not chemically consistent with expectation for spring water. The water chemistry is more consistent with tap water, or spring water that has been treated. The concentrations of total THMs detected in this study are similar to the concentrations in tap water in some Australian locations in 2013/2014,(SA Water 2014; Water Corporation 2014) although it is also possible that tap water with higher THM concentrations was inadequately purified, or that spring water was contaminated during the bottling processes. Nonetheless, the implication of our finding is that researchers cannot assume that bottled water marketed as 'spring' or 'mineral' water in Australia is THM-free as one would expect. Estimates of total THM exposure if water marketed as 'spring' or 'mineral' water was assumed to be THM-free would be under-estimated by up to 30 µg/L. While this is still a relatively small difference, it could result in exposure misclassification of study participants, particularly where bottled water is their primary source of drinking water.

Table 1: Concentrations of trihalomethanes for each brand of bottled water tested

	Concentration (µg/L)														
	Bromodichloromethane			**Bromoform**			**Chloroform**			**Dibromochloromethane**			**Total trihalomethanes**		
Bottle brand	**Median (interquartile range)**	**Range**	**Number of positive bottles[a]**	**Median (interquartile range)**	**Range**	**Number of positive bottles[a]**	**Median (interquartile range)**	**Range**	**Number of positive bottles[a]**	**Median (interquartile range)**	**Range**	**Number of positive bottles[a]**	**Median (interquartile range)**	**Range**	**Number of positive bottles[a]**
A	6 (6-6)	5-6	10	13 (13-13)	12-13	10	2 (2-2)	2-2	10	6 (6-7)	4-7	10	27 (27-27)	23-28	10
B	0 (0-6)	0-7	4	10 (10-11)	0-14	8	0 (0-2)	0-4	4	0 (0-7)	0-7	4	10 (10-26)	0-30	6
C	0 (0-0)	0-0	0	0 (0-0)	0-0	0	0 (0-0)	0-2	1	0 (0-0)	0-0	0	0 (0-0)	0-0	0
D	0 (0-0)	0-0	0	0 (0-0)	0-0	0	0 (0-0)	0-0	0	0 (0-0)	0-0	0	0 (0-0)	0-0	0
E	0 (0-0)	0-0	0	0 (0-0)	0-0	0	0 (0-0)	0-0	0	0 (0-0)	0-0	0	0 (0-0)	0-0	0
F	0 (0-0)	0-0	0	0 (0-0)	0-0	0	0 (0-0)	0-0	0	0 (0-0)	0-0	0	0 (0-0)	0-0	0
Total	**0 (0-0)**	**0-7**	**14**	**0 (0-10)**	**0-14**	**18**	**0 (0-1)**	**0-4**	**15**	**0 (0-0)**	**0-7**	**14**	**0 (0-10)**	**0-30**	**16**

Notes: 10 bottles were sampled for each brand of bottled water. Concentrations of <1 µg/L for the individual types of trihalomethanes and <4 µg/L for the total trihalomethanes have been reported as 0 above. [a]Positive bottles were those where the concentrations exceeded the detectable limits (concentrations of <1 µg/L for the individual types of trihalomethanes and <4 µg/L for the total trihalomethanes).[b]Names withheld

Table 2: Concentrations of trihalomethanes for each bottle of brand B water by batch number, with brand A concentration ranges included as a reference

	Concentration (µg/L)				
Batch number	**Bromodichloromethane**	**Bromoform**	**Chloroform**	**Dibromochloromethane**	**Total trihalomethanes**
MTN1 19:08	0	0	0	0	0
MTN1 19:08	0	0	0	0	0
MTN1 04:36 4	0	2	0	0	0
MTN1 04:44 4	0	2	0	0	0
W1 19:38	0	10	0	0	10
W1 19:40	0	10	0	0	10
07:44 WB2 3213	7	12	4	7	30
03:25 3213 2 3213	6	14	2	7	29
08:21 3213 2 3213	6	13	2	7	28
07:00 3213 2 3213	6	11	2	7	26
Supermarket brand (ranges)	**5-6**	**12-13**	**2-2**	**4-7**	**23-28**

Note: The last four batch numbers resemble those of brand A as all end with 3213. Concentrations of <1 µg/L for the individual types of trihalomethanes and <4 µg/L for the total trihalomethanes have been reported as 0 above, as they are below the detectable limits

Table 3: Trihalomethane concentration guidelines for drinking water

	Concentration (μg/L)				
	Bromodichloromethane	**Bromoform**	**Chloroform**	**Dibromochloromethane**	**Total trihalomethanes**
Global: World Health Organization (2017)	60	100	300	100	Refer to the guidelines for further information
Australia: National Health and Medical Research Council (2017)	60	100	300	100	250
International ranges (excluding the Maximum Contaminant Level Goal (United States of America): Jackson et al. (2008)	16-80	80-100	60-300	60-150	25-250

Notes: The National Health and Medical Research Council (2017) guidelines for the specific types of trihalomethanes are taken from the World Health Organization (2017) guidelines. The Maximum Contaminant Level Goals were 0 μg/L for bromodichloromethane, 0 μg/L for bromoform, 70 μg/L for chloroform and 60 μg/L for dibromochloromethane (Jackson et al. 2008).

One of the challenges for estimating THM exposure identified in this study is where results were inconsistent. The slight differences in concentrations for the supermarket brand water are unlikely to be sufficient to alter associations between THM exposure estimates and health outcomes, although the range of concentrations for the beverage company brand were more substantial. The batch numbers and concentrations indicate that brand B may be sourced from and bottled in at least two, maybe three, locations, with one of those likely to be comparable to brand A. The range of concentrations, for example 0-30 μg/L for total THMs, poses some challenges for estimating THM exposures.

5. Conclusion

Our study showed that bottled water in Australia marketed as 'spring' or 'mineral' water cannot be assumed to be THM-free. When estimating THM exposures, toxicologists and environmental epidemiologists should therefore obtain information about the brand(s) of bottled water being consumed, and obtain data on THM concentrations that are brand-specific. In doing so, exposure estimates will be improved, and the evidence for an association between trihalomethane exposure and human health outcomes will be strengthened, ultimately leading to more accurate public health recommendations.

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