Environ Monit Assess (2022) 194: 144
https://doi.org/10.1007/s10661-022-09783-2

Bromate concentrations and pH values in bottled drinking water in Kuwait

Meshari Al-Qallaf · Walid Q. Alali

Received: 21 September 2021 / Accepted: 15 January 2022 / Published online: 4 February 2022

Abstract The study objectives were to assess bromate concentrations in bottled drinking water sold at retail in Kuwait and assess pH values relationship to bromate concentrations. A cross-sectional study was conducted in 2019 where 120 bottled water samples were collected from supermarkets across six governorates in Kuwait. Samples represented local brands that used distilled or mineral water as well as imported brands that used mineral water only. The samples were analyzed for bromate concentrations and pH values. The overall mean bromate concentration was 4.02 µg/L (95% CI: 3.35–4.69) with concentration of 4.45 µg/L in locally distilled water significantly ($P < 0.05$) higher than that in imported mineral water samples (i.e., 1.34 µg/L). The overall bromate percent positive was 41.7% ($n = 120$) with 35.8% in locally distilled samples, significantly higher than that in imported mineral samples (5.8%). None of the local mineral bottled water samples had detectable bromate. Bromate concentrations in our samples were within the international allowable limits of less than 10 µg bromate/L (except for one local distilled sample that contained 14.9 µg bromate/L). The mean pH value was 7.39 (95% CI: 7.33–7.45). There was no significant relationship between pH values and bromate concentrations in our samples. Our findings proved that local and imported bottled water sold at retail in Kuwait was bromate safe.

Keywords Middle East · Desalination · Water safety · Water contamination

M. Al-Qallaf
Quality Health Safety of Work Environment Department, Kuwait Institute for Scientific Research, Safat, Kuwait City, Kuwait

W. Q. Alali (✉)
Department of Epidemiology & Biostatistics, Faculty of Public Health, Kuwait University, Safat, Kuwait City, Kuwait
e-mail: w.alali@ku.edu.kw

Introduction

Freshwater resources are extremely limited in Kuwait due to its desert nature. The country suffers from extremely high temperatures in the summer, low annual rainfalls, limited groundwater wells, and absence of rivers and lakes. The annual rainfall in Kuwait is about 110 mm (Hamoda, 2001). However, the extreme weather conditions lead to a high-water evaporation rate that makes recharging freshwater resources insufficient. Nevertheless, Kuwait has one fresh groundwater (i.e., mineral) reservoir located in the southern part of the country.

Kuwait has been largely dependent on water desalination as a source of water for drinking and everyday use. Because of the concerns with safety, quality, and taste of desalinated (i.e., distilled) water among many residents, bottled water is a popular source of drinking water in Kuwait as well as in the other

Gulf Cooperation Council (GCC) countries. The consumption of bottled drinking water has significantly increased in the past 20 years worldwide and is expected to continue to rise in the future (Aljundi, 2011; Doria, 2006). In Kuwait, the consumption of bottled water has increased from 50.2 to 76.1 L per capita between 1999 and 2004 (Al-Mudhaf et al., 2011; Alsulaili et al., 2015).

Bromate is not naturally found in water (Haiderkadhim, 2017). The presence of bromate is bonded with the availability of bromide (precursor for bromate) in the water sources (Aljundi, 2011; Othman et al., 2010). Bromide can occur naturally in seawater, or it can enter water sources by human activities such as industrial effluent discharges (Radwan et al., 2021; Siddiqui et al., 1995).

Treated water is considered as one of the main sources of bromate intake. This chemical compound can form during the disinfection stage when ozone is used in water treatment or when hypochlorite solution is used to disinfect drinking water (Haag & Holgen, 1983). Bromate has been classified as a possible carcinogen to humans (IARC, 1999; USEPA, 2001). Although bromate lethal dose has not been defined precisely, there are concerns with the presence of bromate in drinking water at any level.

Bromate has been detected at various concentrations in bottled drinking water. For instance, the presence of high levels of bromate in bottled water at retail level in the UK, (Guardian, 2004), United Arab Emirates (Kannan, 2012), and in Kuwait (Aljoun, 2014) have led to recall of the water products. Moreover, in 2007, 600 million gallons of water from Silver Lake and Elysian Reservoirs in Los Angeles (USA) were drained out due to the presence of high concentrations of bromate (68 and 106 µg/L in the lake and reservoirs, respectively) (Helfand, 2007).

Bromate is a problematic compound in both tap and bottled water. Health-related authorities are aware of the unavoidability of bromate formation during water treatment and the health adverse of bromate through chronic exposure. Due to the limited data on bromate in bottled drinking water in Kuwait, the study objectives were to (1) determine bromate concentrations in retail bottled water of different producers (local and imported) and sources (distilled and mineral) in Kuwait and (2)assess the relationship between pH values and bromate concentrations in the water samples. It is anticipated that the study findings will help in risk assessment and policy decisions regarding the safety of bottled drinking water.

Materials and methods

This manuscript is reported according to Strengthening the Reporting of Observational Studies in Epidemiology (STROBE) protocol (von Elm et al., 2014).

Study population and sampling scheme

A cross-sectional study was conducted between February and December 2019 to determine bromate concentration in bottled water samples collected from retail supermarkets in Kuwait. A total of 120 bottled waters were sampled from 12 cooperative society supermarkets across the six governorates of Kuwait (i.e., Kuwait City, Hawalli, Farwaniya, Jahra, Mubarak Al-Kabeer, and Ahmadi). Two types of drinking bottled waters were included in this study: locally manufactured and imported. Two supermarkets were chosen randomly from a list of all supermarkets per each governorate. The bottled water brands included in the study sampling plan were the most selling brands in Kuwait based on a review of the financial sale records of the supermarkets during the study period. The bottled water samples were collected from three locally manufactured brands and two imported brands. All bottled water brands used desalinated (i.e., distilled) water except for one local manufacturer that used mineral water (i.e., natural mineral groundwater). The distilled water goes through extra purifications via either ozone or ultraviolet (UV) treatment at the water processing plant before filling the bottles.

Samples (whole bottled water of 330–500 mL volume) were collected using a stratified random sampling method where the brand was considered as the stratum. Within each stratum, samples were collected randomly using systematic random sampling. The selected bottles were matched by manufacturing brand, date, and volume.

Upon sample collection, all water samples were transferred into individual sterile 1-L polyethylene bottles (Kuwait national plastic, Sabhan industrial area, Kuwait). The polyethylene bottles were labeled with the date of collection and a code to ensure that samples were "blinded" from the laboratory staff during testing to eliminate bias associated with water brands. The samples were stored at 4 °C in an icebox and transferred to the Research Sector Projects Unit (RSPU) Laboratory in the Faculty of Science at Kuwait University (Khaldiya, Kuwait) for pH and bromate chemical analyses.

pH analysis

Within 24 h from receiving the samples, the pH was measured using METROHM-781 pH/Ion Meter (Metrohm AG, Herisau, Switzerland). The instrument was initially calibrated using standard buffer solutions of pH 7, 4, and 9 per manufacturer's instructions before measuring the water samples. A 100 mL aliquot of each bottled water sample was used for the pH measurements. The pH readings were conducted at ambient temperature. Each sample was measured three times, and the readings were averaged. In addition, the pH value written on the sampled bottled water ingredient label was also recorded.

Bromate analysis

Extraction and analysis of bromate

Similarly, within 24 h from receiving the samples, bromate concentration was measured according to United States Environmental Protection Agency 326.0–1 method (USEPA, 2002) using 881 Compact Ion Chromatography pro, 919 Ion Chromatography Autosampler Plus, and 887 Professional UV–VIS Detector (Metrohm, Switzerland). Each water sample was injected into the instrument at 20 μL volume. The data were obtained using the MagIC Net Software (Metrohm, Herisau, Switzerland). The step of bromate separation was done using Metrosep Column A Supp 16–100/4.0 (Metrohm, Switzerland) followed by post-column reaction step through the triiodide method which was then measured by UV absorption at 352 nm. All bottled drinking water samples will be analyzed directly without any pretreatment steps. Figure 1 demonstrates chromatograms of one of the bromate contaminated sample and for a non-contaminated bottled drinking water sample.

Quality assurance and quality control for bromate analysis

A seven-point calibration ranging from 0.5 to 40 μg/L was performed before the analysis as shown in Fig. 1. The R^2 calibration was 0.996 which is considered acceptable. Instrumental calibration curves were generated at least weekly, and continuing calibration checks were performed daily before and after every batch of 10 samples using a midpoint calibration standard to check for instrumental drift in response. The detection limit was 1 μg/L. A positive control was used by spiking samples with internal laboratory standards for bromate to verify the analysis results. Furthermore, appropriate dilutions of stock positive standards were prepared and analyzed before and after each set of ten samples.

Statistical analysis

Descriptive statistics of bromate concentrations and pH values (mean and 95% confidence intervals) were provided by water origin (local or imported) and source (distilled or mineral). Additionally, we provided bromate percent positive water samples (i.e., number of positive bromate samples at any concentration out of the total number of samples tested) cross-tabulated by water origin (local or imported) and source (distilled or mineral). The relationship between bromate concentrations and the study variables (water origin, water source, and pH values) was accessed via a multiple linear regression model. The proportion (i.e., percent positive) of bromate in water was compared across the levels of each variable (water origin and source) using 2×2 chi-square or 2×n likelihood ratio chi-square test, as appropriate. The relationship between the pH readings from the laboratory and that from the bottles' ingredient labels was assessed using Spearman's rank correlation coefficient. All data analysis was performed using STATA software version 15.1 (Stata Corp., College Station, Texas).

Results

Bromate presence and concentration

The bromate concentration (mean and 95% CI) and the percent positive samples by water origin and

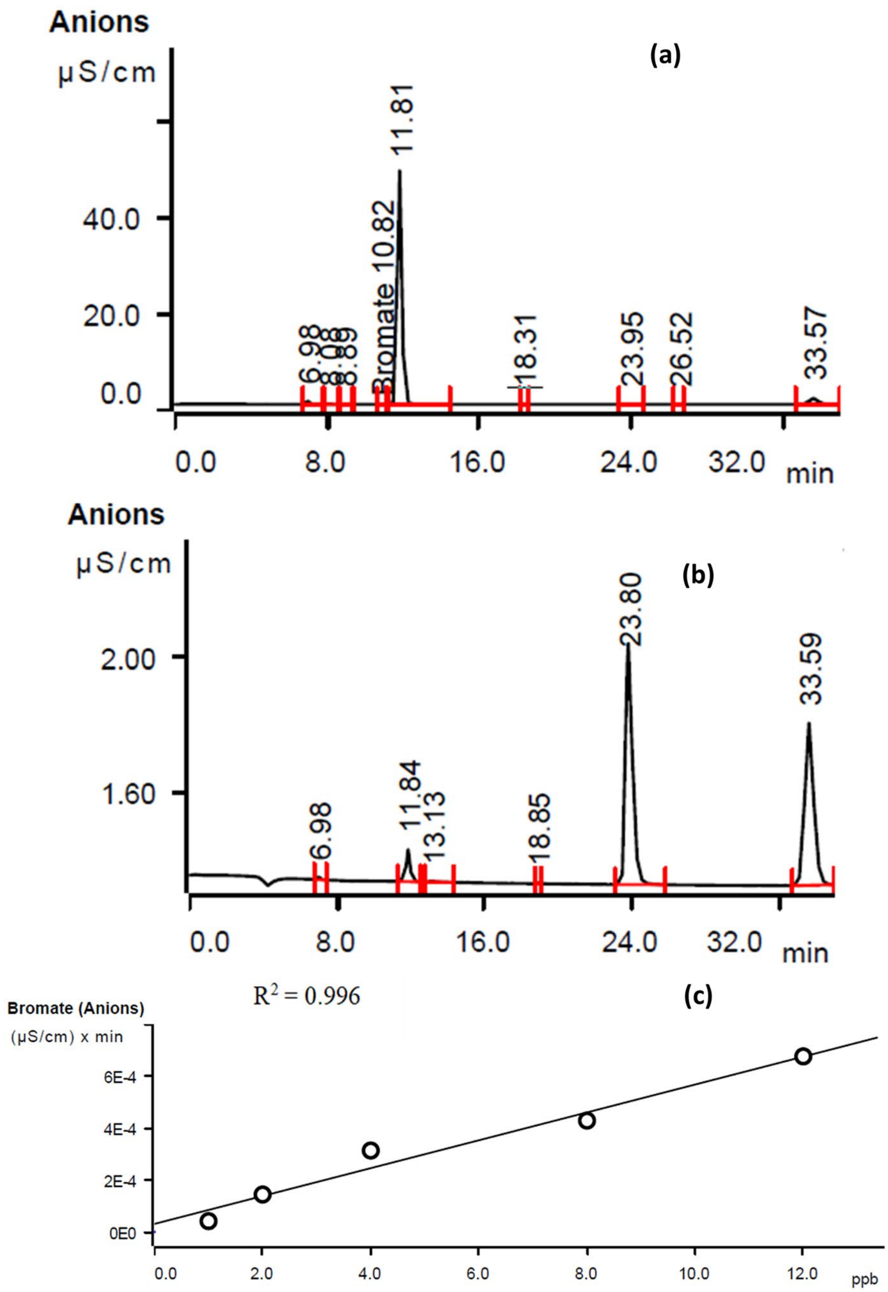

Fig. 1 Chromatograms of **a** one bromate contaminated sample and **b** a non-contaminated sample of bottled drinking water in Kuwait. **c** a calibration curve used with some of the samples ($R^2 = 0.996$)

source are shown in Table 1. The distribution of the bromate concentrations by water origin is shown in Fig. 2. The mean concentration of bromate in locally manufactured distilled water samples (4.45 μg/L [95% CI: 3.8–5.1]) was significantly ($P < 0.05$) higher than that in imported mineral water samples (1.34 μg/L [95% CI: 0.7–2.0]). The overall bromate percent positive was 41.7% ($n = 120$). Moreover, the percent positive in locally manufactured distilled water samples (35.8%) was significantly higher ($P < 0.05$) than that in imported mineral samples (5.8%). None of the locally manufactured mineral bottled water samples had detectable bromate. Although bromate was detected in our samples, the concentrations were under the maximum contamination level (MCL) of 10 μg/L except for one local distilled water sample

Table 1 Bromate concentrations (mean and 95% CI) and percent positive by water origin and source (local/imported and distilled/mineral) based on 120 bottled water samples collected from supermarkets in Kuwait

Water origin and source	Number of samples	No. of bromate positive sample	Mean (µg/L) and 95% CI	% bromate positive
Local distilled	48	43	4.45 (3.8–5.1)	35.8
Local mineral	24	0	0	0
Imported mineral	48	7	1.34 (0.7–2.0)	5.8
Total	120	50	4.0 (3.4–4.7)	41.6

that contained 14.9 µg bromate/L. When comparing the bromate concentrations by location (supermarkets across the six governorates), there were no significant differences between the means or between the bromate percent positives ($P > 0.05$) by location.

Bottled water pH values

The mean and 95% CI of the pH values obtained from the bottled water ingredient label and those obtained from measurements at the laboratory via pH meter are shown in Table 2. There was no significant difference ($P > 0.05$) between the means of the laboratory pH values and those printed on the ingredient labels. All the pH values were within Kuwait Environmental Public Authority (KUEPA) and World Health Organization (WHO) acceptable ranges (i.e., 6.5–8.5) (WHO, 2017). The relationship between bromate concentrations and laboratory measured pH values was not statistically significant ($P > 0.05$). Figure 3 shows the relationship between laboratory measured pH values and bromate concentration.

Discussion

Our findings revealed that bromate concentrations in bottled water sold in Kuwait retail supermarkets are within the international MCL acceptable limits. Alomirah (2019) reported a lower bromate concentration in bottled water (i.e., 2.89 µg/L) from Kuwait compared to our findings in distilled water samples. However, their findings were based only on 10 bromate positive samples out of 19 bottled water tested compared to the 120 samples tested in this study. Moreover, the author did not describe the sampling methods used to collect/choose the water bottles, or provided information on the source of samples (locally produced/imported), or the source of the water (distilled/mineral). In another study by Al-Mudhaf et al.

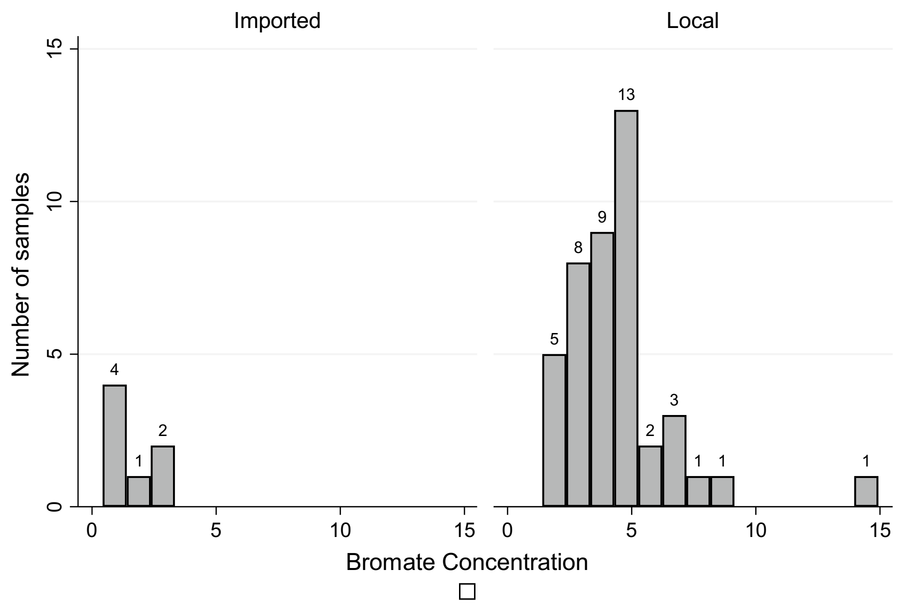

Fig. 2 The distribution of bromate concentrations (µg/L) by water origin

Table 2 The mean and 95% CI for pH values obtained from bottled water ingredient label and those measured in the laboratory based on 120 water samples collected from supermarkets in Kuwait

Bottled water origin	No. of samples	Label ingredient pH	Laboratory measured pH
Local	72	7.2 (7.2–7.4)	7.3 (7.2–7.3)
Imported	48	7.2 (7.1–7.3)	7.5 (7.4–7.6)
Total	120	7.2 (7.3–7.4)	7.4 (7.2–7.3)

(2011), authors revealed that 15 of 71 bottled water samples (26.7%) collected from retail markets had high bromate concentrations (range between 50 and 2400 µg/L). However, their sampling plan did not include different bottled water sources (local and imported) nor it explained their sample selection method from the retail markets. The number of studies from different countries has reported bromate concentrations in drinking water. For instance, in Sudan, bromate was detected in five bottled water samples at concentrations ranging between 18.2 and 169 µg/L (Musa et al., 2010). In a study from India, bromate concentration was above the international MCL of 10 µg/L in 13 bottled drinking water out of 31 samples (Kumar et al., 2011). In Saudi Arabia, Alsohaimi (2012) stated that 17% of the bottled drinking water samples were above the international MCL.

Bromate concentrations in drinking water correlate with the amount of bromide, which is a precursor for bromate, in the water source (Aljundi, 2011; Othman et al., 2010). For instance, seawater has more bromide than freshwater (Alomirah et al., 2019). In the matter of fact, the average bromide concentration in seawater was around 67×10^3 µg/L, whereas the range in freshwater groundwater was 10–1000 µg/L (Alomirah et al., 2019). While some studies have shown that bromate concentrations in water from freshwater sources are usually low, others showed the contrary. This variation is likely due to the sampling methods, number of samples collected, treatment methods, and laboratory analysis methods. For instance, Espino and Cimatu (2003) reported that bromate was not detected in five bottled water from a freshwater source which is in agreement with our study where the 24 local mineral bottled water samples had undetectable bromate concentration. Moreover, in Al-Mudhaf et al. (2011) study, out of 71 bottled water samples (56 natural mineral samples, 13 purified water, and 2 spring water samples) that were collected from

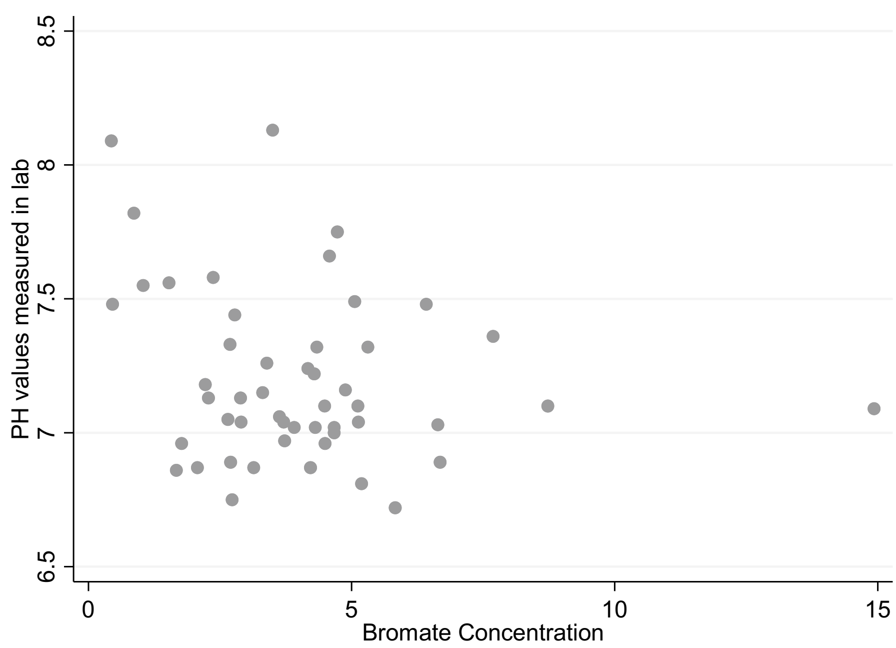

Fig. 3 Relationship between bromate concentrations in (µg/L) and pH lab values

different retail markets in Kuwait, 21.1% was bromate-positive (13 minerals, 1 purified, and 1 spring water sample). The authors found that the highest bromate concentration was in one of the natural mineral water sources (2400 µg/L), which is 240-folds higher than the maximum contamination level (MCL) of 10 µg/L.

The pH is considered as an important operational water-quality parameter. Although pH has generally no direct effect on human health (Al-Mudhaf et al., 2011), health and quality organizations have standardized the pH ranges in bottled water. Furthermore, many customers check the pH value in addition to the sodium content. Bottled water-producing companies are obliged to print the pH value on the container label where consumers can see and read clearly. Furthermore, bottled water companies do not measure the pH for every single bottle they produced; hence, the pH value on the label is based on the quality control testing of some water samples during production. Additionally, the companies use the same printed label for their large daily production of water bottles. Our findings revealed that pH values on bottled water samples and the measured pH values in the laboratory were not significantly different. Additionally, both were within the acceptable pH ranges. This is in disagreement with previous studies that reported measured pH values in bottled water samples were different from the manufacturer ingredient labels (Alsulaili et al., 2015; Khan & Chohan, 2010). The authors of these studies did not report if the difference between the measured pH values and those printed on the ingredient labels was significantly different, whereas in our study, we performed a statistical test to check for significant differences.

The relationship between bromate concentrations and the laboratory measured pH values was not statistically significant in this study. That may indicate that pH values did not have an impact on bromate concentrations in our samples. This is in disagreement with other studies that reported a positive relationship between pH and bromate concentration in bottled water (Aljundi, 2011; Sharif, 2014; Siddiqui et al., 1995). In those studies, lowering pH may have reduced bromate concentrations.

Study limitations

This study has several limitations. First, this is a cross-sectional study; therefore, a temporal association cannot be established between the exposures and outcome. Second, we did not measure the bromide that is naturally present in all water sources. This is because of the unavailability of the laboratory reagents and equipment needed to be determine bromide concentrations. Third, while all bottled water-producing companies in Kuwait use ozone to disinfect the water, we did not have information on the concentration of ozone used, ozone reaction time, or pH value range during the disinfection process. Fourth, we did not measure the storage temperature of the bottled water at sampling to assess its potential impact on bromate concentration. For instance, higher storage temperatures might increase bromate concentration in bottled water. However, in Kuwait, all bottled water must be stored and transported under temperature-controlled conditions. Therefore, we did not anticipate that temperature had impacted bromate concentrations in our samples.

Conclusions

Bromate concentrations in locally manufactured distilled bottled water were significantly higher than that in mineral bottled water (local and imported); however, the concentrations were within the international allowable limits. Additionally, the pH of the water samples did not have a significant impact on bromate concentrations. Moreover, pH values that were measured in the samples (average of 7.4) were not significantly different than those printed on the ingredient label (average of 7.2). Hence, the pH was within the acceptable local and international pH ranges. The most selling locally manufactured (distilled and mineral) and imported (mineral) brands sampled from cooperative society supermarkets in Kuwait during the study period were bromate-safe to drink.

Acknowledgements The authors would like to thank the staff at the Research Sector Projects Unit (RSPU) Laboratory in the Faculty of Science at Kuwait University.

Author contribution MAQ and WQA: study design, sample collection and chemical analysis, data analysis, interpretation, and write-up.

Data availability The data will be available upon request to the corresponding author.

Declarations

Ethics approval and consent to participate The study was approved by the Ministry of Health Standing Committee for the Coordination of Health and Medical Research (i.e., Ethics Committee), Kuwait City, Kuwait (approval number 2019/1087).

Consent for publication Not applicable.

Conflict of interest The authors declare no competing interests.

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