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Original Article

Content of cariostatic trace elements (F, Cu, and Zn) in the commercially packaged and public tap waters of Qatar

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A R T I C L E I N F O

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A B S T R A C T

Objectives: Drinking water containing optimal levels of cariostatic trace elements is an effective preventive approach against community-wide dental caries. This study aimed to (i) determine the concentrations of fluoride, copper, and zinc in commercially packaged and public tap water available in Qatar, (ii) assess the accuracy of the content of these cariostatic trace elements on commercial water brand labels, and (iii) check for seasonal fluctuations in the levels of these trace elements in commercial and public water samples.

Methods: Duplicate samples from 20 commercial and six public tap water sources were collected once in the summer and winter seasons. Water samples were evaluated for fluoride concentration using ion chromatography (IC), whereas copper and zinc concentrations were estimated using inductively coupled plasma mass spectrometry (ICP-MS).

Results: The fluoride content of all commercial and public water samples was below the 0.25 mg/L detection limit of the IC instrument. Commercial water brands had copper and zinc levels below the ICP-MS detection limits (1.22 μg/L for copper and 0.24 μg/L for zinc). Copper levels in the public water samples ranged from <1.22 μg/L to 11 μg/L. Zinc levels in the public water samples ranged from 5 μg/L to 40 μg/L. Thirteen commercial water brands labelled their fluoride content, with most showing labelled fluoride levels <0.1 mg/L, consistent with the results of this study. The difference in trace element content between the summer and winter water samples was not statistically significant ($P > 0.05$).

Conclusions: Lower optimal cariostatic trace element content in the drinking water in Qatar could be a reason for the exceptionally high prevalence of dental caries. The results of this study could lead to public health advocacy for supplementing drinking water in Qatar with cariostatic trace elements that could potentially lower the prevalence of dental caries.

1. Introduction

The most recent Qatar National Oral Health Surveys indicate that the prevalence of dental caries in Qatar are among the highest in the world, ranging from 72 % in 6-year-old children to 85 % in 12–14-year-olds, and reaching almost 90 % in preschool children (Chrisopoulos et al., 2024; Al-Darwish et al., 2014; Alkhtib et al., 2016; Al-Thani et al., 2018). The high dental caries rates in Qatar have been attributed to the frequent intake of dietary sugars and lack of awareness of oral health (Bener et al., 2013). There is a need for intensive public health dental caries prevention programs to lower disease prevalence rates in Qatar. Drinking water containing cariostatic trace elements, such as fluoride, copper, or zinc, can be a particularly attractive caries preventive approach as it is universally consumed by the entire population.

The cariostatic effects of fluoride are well established and are mediated predominantly through its topical influence on the tooth surface demineralisation-remineralisation equilibrium (Buzalaf et al., 2011). The significant reduction in the prevalence of caries in developed countries has been attributed to the addition of fluoride to public drinking water supplies and dentifrices (Fejerskov 2004; Murthy 2015). The potential role of other trace elements, such as copper and zinc, in reducing dental caries has also attracted research interest. Clinical studies have demonstrated that rinsing with aqueous solutions containing low concentrations of copper and zinc results in significant

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synergistic inhibition of dental plaque acidogenicity and enamel demineralisation (Afseth et al., 1980; Afseth 1983; Afseth et al., 1983; Abdullah et al., 2006). The salivary concentrations of copper and zinc have shown a consistent inverse relationship with dental caries (Duggal et al., 1991; Hussein et al., 2013; Hegde et al., 2014; Bhandary et al., 2015; Hussein et al., 2017). A recent systematic review concluded that the presence of copper and zinc in the oral environment negatively affected the dental caries process (Khan et al., 2020).

Effective and safe dental caries prevention programs require awareness of the precise concentration of cariostatic trace elements in drinking water consumed by the general public. The optimal fluoride level in drinking water is the concentration at which substantial caries reduction can be achieved while limiting the risk of dental fluorosis. The U.S Public Health Service recently lowered the recommended fluoride concentration in drinking water to 0.7 mg/L to balance the risk of dental fluorosis with protection against dental caries (USPHS 2015). However, since people living in hot climates like Qatar drink more water, recommended fluoride levels in drinking water are even lower, in the range of 0.6–0.7 mg/L (Aldosari et al., 2003; O'Mullane et al., 2016). The optimal concentrations of copper and zinc in drinking water for prevention of caries have not yet been determined. The World Health Organization (WHO) guidelines recommend 2 mg/L of copper in drinking water to avoid metallic taste and toxicity issues (WHO 2004). The recommended daily allowance (RDA) for copper in adults is 900 μ g/day. The WHO has not set a guideline value for zinc in drinking water, but levels above 3 mg/L tend to give water an astringent taste (WHO 2003). The RDA for zinc in adults is 8–11 mg/day.

The two major sources of drinking water in Qatar are (i) the public supply of desalinated seawater and (ii) commercially packaged water available in bottles or gallon containers. It is estimated that only 30 % of the population in Qatar drinks public tap water supplied by the Qatar General Water and Electricity Corporation, although public tap water continues to be used for cooking in most households. (QEERI 2015). Following global trends, Qatar has witnessed an increase in the consumption of commercially packaged drinking water because of the perception that it is healthier and devoid of chemicals and harmful microbes. However, studies worldwide have shown that the fluoride content of commercially packaged water is lower than the recommended level for dental caries prevention (Supplementary Table 1).

A 2016 study in Qatar found wide variations in the fluoride levels for different brands of commercial bottled water supplied to the country (Almulla et al., 2016). Moreover, many commercial bottled water manufacturers do not mention the concentrations of essential trace elements on the outer labels of their products, and if mentioned, these levels may not necessarily be accurate (Venturini and Frazão 2015). A large majority of the bottled water brands evaluated in the 2016 study are no longer available in the country, and there are no contemporary reports on the levels of fluoride in packaged drinking water or public tap water in Qatar. Additionally, to the best of our knowledge, there are no reports on the availability or concentration of other potential cariostatic elements, such as copper and zinc, in the water supplies of Qatar. Therefore, this study aimed to (i) determine the concentrations of fluoride, copper, and zinc in the commercially packaged and public tap waters of Qatar, (ii) confirm the accuracy of any labelling of cariostatic trace element content in commercial water packages, and (iii) determine whether seasonal fluctuations (summer vs. winter) influence the availability of these cariostatic trace elements in drinking water.

2. Materials and methods

2.1. Water collection

Twenty commercially packaged drinking water brands were purchased from major supermarkets in Qatar and unfiltered public tap water samples were collected from six geographical locations in the country (Supplementary Table 2). Commercial water brands included 14 brands of bottled drinking water and six brands of drinking water stored in gallon containers. Two samples were obtained from each commercial brand, each with a different batch number and bottling date. The brand label information on fluoride, copper, and zinc levels was recorded. Both commercial and public water samples were collected twice, once in the summer (July or August) and once in the winter (January or February), for separate experimental analyses. All water samples were stored in their original closed plastic containers at room temperature in the dark until the trace element analyses were performed.

2.2. Trace element analysis

The fluoride concentrations of the water samples collected were analysed by ion chromatography (IC), based on the standard EPA 300.1 methodology (EPA 1997). The 850C Professional IC system (Metrohm, Herisau, Switzerland) employed in this study has a lower fluoride detection limit of 0.25 mg/L. Water samples were injected directly into the IC system without dilution and eluted from the anion exchange column and electroconductivity detector. Anion identification was determined by retention time and quantitation by comparing the conductivity peak areas of the samples with those of accurately prepared analytical standards. Copper and zinc levels were estimated using an inductively coupled plasma mass spectrometer (ICP-MS), based on the standard EPA 200.8 methodology (EPA 1994). A NexION™ 300D ICP-MS (PerkinElmer, Shelton, CT, U.S.A) was used to determine the copper and zinc concentrations. The ICP-MS used in this study had lower detection limits of 1.22 μg/L for copper and 0.24 μg/L for zinc. Two readings were taken for the fluoride, copper, and zinc levels in each water sample tested, and the average was recorded. Trace element analysis was performed separately for the water samples collected during the summer and winter months.

2.3. Statistical analysis

The Statistical Package for the Social Sciences v22 (SPSS) was used to derive descriptive data. Paired t-tests and correlation analyses were employed to compare the first and second measurements of the water samples, between different batches of water samples, and between water samples collected in the summer and winter seasons.

3. Results

3.1. Trace element content in commercial packaged water samples

The measured fluoride, copper, and zinc contents of the 20 commercial drinking water brands tested in this study are listed in Table 1. The accuracy of the experimental methods was confirmed by comparing the first and second readings of the samples; the differences were not statistically significant (P=0.99). The correlation between the two readings was $r = 0.998$ ($P < 0.0001$). The differences in trace element contents between the two batches of commercial water brands and between the summer and winter water samples were not statistically significant (P>0.05). All commercial water brands contained trace element concentrations below their respective detection limits (0.25 mg/L for fluoride, 1.22 μg/L for copper, and 0.24 μg/L for zinc). These results were consistent for water samples collected during the summer and winter months.

Regarding the labelling of trace elements in commercially packaged drinking water, no brand specified their copper or zinc contents. For fluoride labelling, 13 of the 14 bottled water brands displayed fluoride content, although none of the six-gallon water brands labelled their fluoride content. Nine of the 13 labelled fluoride concentrations on bottles were in the range of 0–0.1 mg/L, and these values were in agreement with this study's measurements. The Aquafina® and Qatar Oasis® brands labelled their fluoride content as 0.5 mg/L and 0.43 mg/L, respectively, but both were found to contain < 0.25 mg/L fluoride in

Table 1
Fluoride, Copper, and Zinc content of commercial drinking water brands available in Qatar.

Commercial Brand (Package)	Labelled F content (mg/L)	Measured Mean F content (mg/L)		Measured Mean Cu content (μ g/L)		Measured Mean Zn content (μ g/L)	
		Summer samples	Winter samples	Summer samples	Winter samples	Summer samples	Winter samples
Alkalive (Bottle)	<0.01	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Aquafina (Bottle)	0.5	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Aqua Gulf (Bottle)	<1.0	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Aqua Panna (Bottle)	<0.1	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Arwa (Bottle)	<0.1	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Aseel (Bottle)	<0.1	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Alshamal (Bottle)	1.0	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Evian (Bottle)	0.06	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Fiji (Bottle)	0	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Jouf (Bottle)	<0.1	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Qatar Oasis (Bottle)	0.43	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Rayyan (Bottle)	<1.0	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Tannourine (Bottle)	Not specified	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Voss (Bottle)	0.1	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Doha (Gallon)	Not specified	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Marwa (Gallon)	Not specified	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Nice (Gallon)	Not specified	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Safa (Gallon)	Not specified	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Sabeel (Gallon)	Not specified	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24
Wataniya (Gallon)	Not specified	<0.25	<0.25	<1.22	<1.22	<0.24	<0.24

this study's measurements. Two bottled water brands (Rayyan® and Aqua Gulf®) labelled their fluoride content as < 1.0 mg/L; however, this study found their fluoride concentration to be < 0.25 mg/L.

3.2. Trace element content in public water samples

The fluoride, copper, and zinc concentrations in public tap water sourced from six geographical locations in Qatar are presented in Table 2. The fluoride content of all public water sources was below the 0.25 mg/L detection limit in both the summer and winter water samples. Copper levels in the public water samples ranged from < 1.22 μg/L in the Airport area to 11 μg/L in the Al-Gharrafa area. All public water sources had zinc content above the detection limit and ranged from 5 μg/L in the Bani Hajer area to 40 μg/L in the Al-Dayeen area.

4. Discussion

Drinking water, whether consumed directly or added to foods and beverages, is an important source of essential trace elements required for oral and dental health. The benefits of optimal water fluoridation for dental caries reduction are well-documented and have been shown to consistently decrease dental caries by over 25 % at the population level (Zokaie and Pollick 2022). The cariostatic effects of trace elements, such as copper and zinc, are also attracting increasing attention (Khan et al., 2020), along with their established roles in the prevention and management of oral conditions such as recurrent aphthous stomatitis (Halboub et al., 2021). Therefore, determining the content of these beneficial trace elements in drinking water is essential for establishing an effective dental caries prevention regimen, both at the individual and population levels. While multiple studies worldwide have reported public and commercial drinking water fluoride levels, this is the first study to evaluate the concentrations of all three cariostatic trace elements (fluoride, copper, and zinc) in both commercial and public tap water supplied to a country.

The fluoride contents of the six public water sources tested in this study were below the levels required for dental caries prevention. This could be due to the fact that desalinated seawater is the main source of public water in Qatar. While natural seawater contains approximately 0.7–1.3 mg/L fluoride, the process of desalination reduces fluoride to very low levels. The Qatar General Water and Electricity Corporation has set the maximum permissible level of fluoride in drinking water to be 1.5 mg/L based on guidelines set by the WHO. However, the supplementation of desalinated seawater with essential trace elements before it is publicly distributed is unlikely. Community water fluoridation is a population health measure that is in a unique position to equitably prevent dental caries across all socioeconomic groups (Zokaie and Pollick 2022). Drinking fluoridated water or ingesting meals prepared with fluoridated water is the simplest way to maintain a constant low dose of fluoride in the oral environment, thereby providing significant protection against dental caries (Cury and Tenuta 2008). Reviews by the U.S. Community Preventive Services Task Force, the U.K. Medical Research Council, and the Australia National Health and Medical Research Council have confirmed that fluoridation of community drinking water significantly decreases dental caries rates among children, and stopping water fluoridation increases caries levels (McDonagh et al., 2000; Truman et al., 2002; Yeung 2008). Adding an appropriate amount of fluoride to the public water supplied in Qatar could be a cost-effective caries-preventive measure that will help significantly lower the high dental caries burden observed among children in the country.

Regardless of the quality and content of public tap water supplied in

Table 2
Fluoride, Copper, and Zinc content of public tap water supplied in Qatar.

Public Water Source	Measured F content (mg/L)		Measured Cu content (μ g/L)		Measured Zn content (μ g/L)	
	Summer samples	*Winter samples*	*Summer samples*	*Winter samples*	*Summer samples*	*Winter samples*
Al-Dayeen Area	<0.25	<0.25	1.81	1.80	40.0	33.0
Al-Gharrafa Area	<0.25	<0.25	11.0	8.0	20.0	15.0
Airport Area	<0.25	<0.25	<1.22	1.6	13.0	9.0
Al-Shamal Area	<0.25	<0.25	1.90	2.7	11.7	11.0
Al-Wakrah Area	<0.25	<0.25	<1.22	1.5	37.0	34.0
Bani Hajer Area	<0.25	<0.25	4.0	2.7	5.0	5.0

Qatar, a large majority of the country's residents prefer commercially packaged drinking water because of the perception that it is healthier (Almulla et al., 2016). The measured fluoride content of all commercial brands assayed in this study was below the recommended levels for dental caries prevention. The fluoride content of commercial water brands in Qatar is consistent with other global studies that found that bottled water contains less than the optimum fluoride concentration required for dental caries prevention (Zohouri et al., 2003; Jimenez-Farfan et al., 2004; Ahiropoulos 2006; Cochrane et al., 2006; Aldrees and Al-Manea 2010; Mills et al., 2010; Dhanuthai and Thangpisityotin 2011; Moslemi et al., 2011; Somasundaram et al., 2015; Almulla et al., 2016; Walia et al., 2017). Except for the Rayyan®, Fiji®, and Evian® water brands, which are sourced from natural groundwater or aquifers, other commercial water brands supplied in Qatar do not clearly state the source of their water. It is likely that most commercial water manufacturers in Qatar source their water from desalinated seawater, resulting in low fluoride content. Commercial water manufacturers are also likely to use distillation and reverse osmosis filtration systems to purify water, which are known to remove fluoride from water at a high rate (Jobson et al., 2000).

Only 13 of the 20 commercial drinking water brands evaluated in this study labelled their fluoride content. Our findings indicate that nine out of the 13 commercial brands showed accurate fluoride content values on their labels, with most showing labelled fluoride levels of $<$ 0.1 mg/L, matching the results of this study. Other studies have reported a significant disparity between the manufacturers' labelling of fluoride content and measured levels (Ahiropoulos 2006; Aldrees and Al-Manea 2010; Almulla et al., 2016). Two bottled water brands (Rayyan® and Aqua Gulf®) labelled their fluoride content as $<$ 1.0 mg/L. However, such range labelling is of limited value in determining suitability for caries prevention, even if the actual fluoride level is within the recommended range (Almulla et al., 2016). This study also reported no differences in fluoride concentrations among different batches of the same commercial brand. These results disagree with reports of significant differences in fluoride concentrations between batches of commercial water products (Bartels et al., 2000; Quock and Chan 2009; Mills et al., 2010), but are in agreement with the results of a Saudi Arabian study which showed no differences in fluoride content between different batches of the same commercial water brand (Aldrees and Al-Manea 2010).

The copper and zinc levels in public and commercially packaged water supplied in Qatar were also very low. All commercial water brands had copper and zinc levels below the instrument detection limits for these elements. Public water samples from different regions in the country showed some variation in the levels of copper and zinc, but were all significantly below the RDA for these trace elements. Unlike fluoride, the cariostatic roles of copper and zinc in drinking water have not been clearly established. However, emerging clinical studies have indicated that the consistent presence of low concentrations of copper and zinc in saliva can lower the incidence of dental caries (Duggal et al., 1991; Hussein et al., 2013; Hegde et al., 2014; Bhandary et al., 2015; Hussein et al., 2017). Low concentrations of copper and zinc in the oral environment can inhibit enamel dissolution by stabilising the enamel crystal lattice (Klimuszko et al., 2018). Furthermore, copper may exert cariostatic effects by inhibiting bacterial growth and various bacterial metabolic enzymes (Khan et al., 2020). Despite the nutritional and biochemical essentiality of copper and zinc in maintaining health throughout life, national food surveys have revealed marginally to moderately low contents of both elements in typical diets (Ma and Betts 2000). Adding appropriate amounts of copper and zinc to drinking water could potentially overcome nutritional deficiencies and play a local role in preventing dental caries. However, further research is required to understand the feasibility and acceptability of drinking water supplemented with copper and zinc. It is also vital to establish the optimal levels of copper and zinc in drinking water to prevent caries while avoiding the associated taste and toxicity issues.

The role of seasonal variations in the concentrations of fluoride, copper, and zinc in drinking water was also assessed in this study. Previous studies have indicated seasonal variability in fluoride content of bottled water, with reports of higher fluoride concentrations in warm and dry climates (Grobler and Dreyer 1988; Tate and Chan 1994; Davraz et al., 2008; Moslemi et al., 2011). These fluctuations in fluoride content were speculated to be related to changes in rainfall and groundwater variability (Cochrane et al., 2006). However, this study did not find significant variability in the trace element content between public and commercial water samples collected in the summer and winter months. This could be because most commercial and public water samples were sourced from desalinated seawater, which is unlikely to show seasonal variations. However, the three bottled water samples sourced from the groundwater or aquifers tested in this study did not show any seasonal variability in trace element content. Consistent and stable levels of cariostatic trace elements in water throughout the year are crucial to maintain their long-term beneficial effects.

5. Conclusion

This study adds to the global evidence that commercially packaged bottled water do not contain the optimal fluoride level required for dental caries prevention. Unfortunately, even the public tap water sources tested in this study had fluoride concentrations below the recommended levels for dental caries prevention. The negligible fluoride content in the drinking water supplied in the country could be one of the reasons Qatar faces an exceptionally high caries challenge among its children. Healthcare professionals should strongly advocate for optimal fluoridation of drinking water supplied in the country as one part of an effective dental caries prevention program. Furthermore, public health authorities should consider issuing official regulations to compel packaged water manufacturers to add the appropriate amount of fluoride to prevent caries.

CRediT authorship contribution statement

Hissa Al-Ansari: Data curation, Investigation, Methodology, Resources, Writing – review & editing. **Hend Al-Qashouti:** Data curation, Investigation, Methodology, Resources, Writing – review & editing. **Roudha Al-Neama:** Data curation, Investigation, Methodology, Resources, Writing – review & editing. **Aisha Al-Moadhadi:** Data curation, Investigation, Methodology, Resources, Writing – review & editing. **Hani Nazzal:** Conceptualization, Supervision, Validation, Writing – review & editing. **Nebu Philip:** Conceptualization, Supervision, Validation, Writing – original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Author contributions

Hani Nazzal and Nebu Philip conceived and designed the study. Literature review, material preparation, data collection, and analysis were performed by Hissa Al-Ansari, Hend Al-Qashouti, Roudha Al-Neama, and Aisha Al-Moadhadi. Nebu Philip drafted the manuscript and all authors critically revised the manuscript. All authors have read and approved the final version of the manuscript.

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Appendix A. Supplementary data

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