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Fluoride Concentration of Drinking Water in Babil-Iraq

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Abstract: The role of fluoride in reducing the risk of dental caries is well documented and is the basis for current intake recommendations. The aim of this study was to evaluate the fluoride content of tap and bottled water currently consumed in Babil-Iraq and to determine whether fluoride intakes by Iraqi consumers fell within the recommended ranges. Fluoride concentrations of 50 samples of tap water (originated from the Euphrates River) and forty popular brands of bottled water currently sold in Babil-Iraq were determined using an Ion-selective electrode. The mean fluoride content of tap and bottled water were 0.184±0.041 and 0.073±0.066 mg L^{-1}, respectively. The average volume of water consumed by Iraqi adults daily was estimated to be 800±240 mL in winter to 2000±650 mL in summer. Based on these data the average daily intake of fluoride by Iraqi consumer from tap and bottled water were 0.147±0.055 to 0.368±0.145 mg and 0.058±0.056 to 0.146±0.140 mg, respectively. These levels revealed that whether tap or bottled water are used as the primary source of drinking water, then Iraqi consumers are at a higher risk of tooth decay. Water fluoridation is recommended as a relevant public health measure to increase the resistance to dental caries.

Key words: Babil-Iraq, fluoride, ion-selective electrode, fluoride determination, dental carries

INTRODUCTION

Water, used directly as a drink or indirectly to prepare beverages and foods, is generally, the main source of ingested fluoride (National Research Council (US) Committee on Fluoride in Drinking Water, 2006). A concentration of about 1 mg L^{-1} in drinking water was identified now as being "optimal" both in reducing caries prevalence and keeping dental fluorosis below 10% in the population (Griffin *et al*., 2007; Ferla *et al*., 2010; Hussain *et al*., 2010). From this "optimal" water fluoride concentration derives the estimated adequate fluoride intake of Infants, children and adults.

As fluoride concentrations in drinking water differ between countries and within countries dependent on natural circumstances and on water fluoridation. Then knowledge of the fluoride levels in drinking water is an important issue to all health care professionals especially dentists (Shailaja and Johnson, 2007).

In Iraq the main source used for producing drinking water is surface water, taken from the Euphrates and Tigris rivers and their tributaries. The municipalities are responsible for the production and delivery of drinking water. The production of drinking water involves generally, conventional water treatment processes (coagulation, flocculation and clarification) that remove suspended matter and disinfection to remove biological agents (Barbooti *et al*., 2010). Chlorine is the most widely used and approved disinfectant in Iraq.

Two destructive wars and a decade of debilitating sanctions, have left the Iraqi water infrastructure struggling and for the most part, failing to provide Iraqis with potable water. During 2004 and 2005 the quality of drinking water in Iraq was greatly deteriorated. The failure of chemical and bacteriological analysis reached about 40% of the collected samples (Barbooti *et al*., 2010). These events have resulted in many Iraqis turning to bottled water with a strong believe that bottled water is healthier than tap water and free of impurities. As a result, the consumption of bottled water has increased dramatically over the last six years.

However, this shift to bottled water may be a concern from a dental health perspective. There has been speculation that if the level of fluoride in bottled water is too low, optimal caries prevention will not be achieved whilst if the levels are too high, developing teeth may be at risk of fluorosis.

Previous research attempts in various countries (Dobaradaran *et al*., 2008; Ghaderpoori *et al*., 2009) found that the majority of the commercially available bottled water failed to list the fluoride content. Some studies have shown several top selling brands containing a fluoride content above the recommended level. On the other hand, some products may have low fluoride content in their composition (De Souza *et al*., 2009).

In Iraq, few reports have been published about the fluoride content of tap and bottled water. Only recently Alsudani *et al*. (2009) and Barbooti *et al*. (2010) reported that the fluoride concentration of tap water (originally from Tigris River) collected during 2008 to 2009 from public water plants in Baghdad was between 0.160 and 1.5 mg L^{-1}. None of these studies, however, measured the amounts of fluoride in drinking tap water originated from Euphrates River or in bottled water currently sold in Iraqi markets.

The present study was designed to assess the concentration of fluoride in tap and bottled water currently consumed in Babil-Iraq, as part of a national program to monitor fluoride ingestion.

MATERIALS AND METHODS

The site: The study was undertaken in a province situated on the Euphrates River named Babil (100 km south of Baghdad). The governorate is located on the site of the ancient Babylonian civilization, which dates back to the beginning of the second millennium BC. Babil, with an estimated population of 1, 385, 783 people in 2003 (Coalition Provisional Authority, 2003) is one of five governorates in south of Baghdad (Basrah, Al-Muthanna, Babil, Kerbala, Al-Qadisiya), in which less than half of the households have access to safe and stable drinking water (MPDC, 2005).

Sampling: Fifty samples of drinking tap water (originally from the Euphrates River) were collected for this study. Forty samples were taken from different locations in Babil and the remaining ten were freshly produced from the production stations. None of the water systems in Iraq had fluoridation or defluoridation units; therefore, fluoride concentration of treated water would be approximately equal to fluoride content of raw water.

All the drinking water samples were monitored in June 2010 to May 2011. To account for seasonal variation of fluoride in water, sampling in each site was replicated 3-4 times in various seasons and the mean value was presented. The water samples were collected directly into labeled plastic bottles with pressure caps. The water from the tap was allowed to flow from 1 to 2 min before the bottles were filled.

Of the 40 bottled water tested, 17 brands had their source or production site from the central Iraqi cities (Babil, Baghdad, Najaf and Kerbala), 11 were from north Iraqi cities (Duhok, Zakho, Sulaimanya and Kurkook) and 12 were imported water from Iran, Kuwait, Saudi Arabia, Turkey and Jordan. Bottled water manufactured in Iraq are either: spring water from the mountainous region in the north, or tap water purified by reverse osmosis. The bottles were made of Polyethylene Terphethalate (PET) and presented in the following forms: 300 mL plastic cups, 500 mL bottles and 1.5 L bottles.

The average volume of water consumed by Iraqi adults daily was assessed via two questionnaires among 200 university students (ages 19 to 26 years), one in January to account for winter consumption and one in July to account for summer consumption. The amounts of drinking water and beverages consumed were estimated by measuring the volume in mL, using drinking vessels such as glasses, cups, mugs and bottles.

Instruments and chemicals: The fluoride concentration in water samples was determined with a combination fluoride selective electrode (WTW, model F800), coupled with an ion analyzer (WTW, model inoLab pH/ION 735, GmbH Company). The fluoride ion selective electrode is the most widely used for the determination of F^- due to its simplicity and short analysis time. Prior to fluoride determination, the samples were mixed with equal quantities by volume of TISAB (Total Ionic Strength Adjustment Buffer). The TISAB contains an acetic acid/acetate buffer that fixes the pH of the solution at about 5. At this pH the formation of HF is negligible and the concentration of OH^-, the only other anion that the electrode responds to, is insignificant. It also contains NaCl to establish a high and constant ionic strength and a complexing agent that removes cations that could interfere by forming complexes with fluoride. Fluoride standards ranging from 0.020 to 0.500 mg L^{-1} fluoride were used to calibrate the measurement. The pH and conductivity of the water samples were also measured using a pH-meter (Model 720, WTW) and a conductometer (Model 720, WTW), respectively. Electrical conductivity and pH values were used as basis to measure total soluble salt concentration in drinking water and the activity of hydrogen/hydroxide in water, respectively.

Analytical-reagent grade chemicals (Merck, Darmstadt, Germany) were used without further purification. Doubly distilled water was used throughout the experiments. The glassware was kept overnight in a 5% nitric acid solution prior to being used.

Statistical analysis: Quantification of fluoride concentrations in the samples was carried out by use of the standard addition method. This is the preferred method as the sensitivity of the potentiometric analysis may vary between samples of different ionic strength (Skoog *et al*., 2004). Data were analyzed using the Statistical Package for Social Sciences (SPSS, version 11.0). The Analysis of Variance (ANOVA) was employed when necessary to detect significance differences among means. Unless otherwise indicated, data are presented as Mean±SD. A probability level of $p<0.05$ was considered statistically significant.

The accuracy has been tested using spike recovery approach. Three additions (spikes) to 5 samples produced average recovery of 97.8%. Relative Standard Deviation (RSD) obtained from 10 measurements during one day has been 3.1%. During one year of work, the calibration curve was always linear in the range 0.050-0.500 mg L^{-1} with a slope between 52 and 58 mV/pF which is very close to Nernstian calibration slope.

RESULTS AND DISCUSSION

Table 1 and 2 demonstrate the Fluoride content (mg L^{-1}), pH and EC (µS cm^{-1}) of tap water and forty brands of bottled water samples available in Babil-Iraq.

Electrical Conductivity (EC): The drinking tap water in Babil is characterized by relatively high EC values (923-1384 µS cm^{-1}), probably due to concentrated colloids and dissolved salts in Euphrates River. The EC values for Iraqi bottled water were in the range 84-428 µS cm^{-1} which suggests that bottled water under consideration have low to moderate salinity (Masoud *et al*., 2003).

The TDS values of tap and bottled water in Babil, are perfectly correlated with EC values. The corresponding values fall between 486 and 860 mg L^{-1} and 50 to 270 mg L^{-1}, respectively. There is no WHO guideline values derived for the permissible level of EC or TDS. However, the palatability of water with a TDS level of less than 600 mg L^{-1} is generally considered to be good; drinking-water becomes significantly and increasingly unpalatable at TDS levels greater than about 1200 mg L^{-1} (WHO, 2006, 2008). Based on this criterion, bottled and tap water in Babil, may be considered palatable.

Currently, the water level in the Euphrates is declining and the salinity of the river has increased, especially in the southern governorates (including Babil). This increased salinity of the water is limiting the provision of potable water from cheaper technology and as such, production of water is and will continue to depend heavily on high technology such as reverse osmosis.

Table 1: Fluoride content (mg L^{-1}), pH and EC (µS cm^{-1}) of tap and bottled drinking water samples in Babil-Iraq

Water type	Source of water	Fluoride label information	Average fluoride concentration	Conductivity	pH
Tap water	Babil's Municipality (50 samples)	-	0.184±0.041 (0.129-0.260)	1159±139 (923-1384)	7.75±0.17 (7.54-8.02)
Bottled water*					
Babbete	Babil	<0.1	0.053	289	7.31
Oasis	Babil	NL	0.026	300	7.21
Alghadeer	Babil	NL	0.042	215	7.35
Bikhal	Babil	0.11	0.063	385	7.23
Daly	Babil	0.11	0.035	106	7.14
Nakee	Babil	0.11	0.038	159	7.20
Zamzam	Babil	NL	0.045	220	6.83
Nabaa Alfurat	Babil	0.70	0.056	296	6.92
Aali Dijlah	Baghdad	NL	0.041	172	7.50
Jeema	Baghdad	NL	0.036	84	6.96
Ater Al Nada	Baghdad	NL	0.037	159	6.96
Furat	Baghdad	NL	0.061	274	7.59
Warda	Baghdad	0.02	0.036	118	7.06
Al Farah	Baghdad	NL	0.046	285	7.20
Joud	Baghdad	NL	0.069	207	6.85
Alsaffa	Najaf	NL	0.043	146	7.21
Noor Hayat	Kurkook	NL	0.079	393	8.06
Mina	Kurkook	NL	0.043	202	7.49
Kameran	Kurkook	NL	0.127	248	7.15
Daiana	Kurkook	NL	0.068	317	7.65
Altoon Alhayat	Kurkook	NL	0.077	112	7.67
Lolav	Duhok	0.03	0.101	223	7.45
Mazi	Duhok	NL	0.056	176	7.22
Rama	Duhok	NL	0.071	245	6.88
Rovian	Duhok	<0.11	0.242	285	7.72
Life	Duhok	0.03	0.078	210	7.05
Sulav	Zakho	NL	0.049	185	7.28
Ala	Sulaimanya	NL	0.337	428	7.50
	Total		0.073	230	7.27

NL: Not labeled; *All averages derived from five readings with a maximum standard deviation of 15%

The pH values: The pH values of tap and bottled water samples in Babil lie between 6.83 and 8.06 (Table 1). They are positively correlated with electrical conductivity (r = 0.38).

Until now, no health base guidelines are proposed for the pH of drinking water, but WHO proposes a desirable range of 6.5 to 8.0 (WHO, 2008). Accordingly, the pH values of all the water sources in the present work are within the WHO desirable limit.

Fluoride concentration: In general fluorides of tap and bottled water showed positive correlation with pH (r = 0.344) and electrical conductivity (r = 0.51). The fluoride concentration of Babil's tap water sources was ranged from 0.129 to 0.260 mg L^{-1} with an average level of less than 0.184 mg L^{-1} (Table 1). These low levels are consistent with similar studies performed outside Iraq for unpolluted drinking water. They are usually considered by WHO to be insufficient to prevent caries (WHO, 2002, 2008; Zohouri *et al*., 2003; Mesdaghinia *et al*., 2010; Shams *et al*., 2009; Pehrsson *et al*., 2006; Al-Salamah and Nassar, 2009; Vandevijvere *et al*., 2009). In some countries, particularly parts of Baltic countries, Central Europe, India, China, Central Africa and South America drinking water can contain very high concentrations of naturally occurring fluoride-well in excess of the WHO's recommended Guideline Value of 1.5 mg L^{-1} (Indermitte *et al*., 2009; Ayoob and Gupta, 2006; Fordyce *et al*., 2007; WHO, 2008).

The mean fluoride content of the 28 bottled water manufactured in Iraq was 0.073 mg L^{-1} with a range from 0.026 to 0.337 mg L^{-1} (Table 1). The highest mean fluoride concentration was found in Ala which contained 0.337 mg L^{-1}, followed by Rovian which contained 0.242 mg L^{-1}. The lowest concentration of fluoride was found in Oasis (0.026 mg L^{-1}) and Daly (0.035 mg L^{-1}) from Babil. These levels were expected for bottled water in Iraq since reverse osmosis technology (currently employed in Iraq) is well known effective method for the reduction of fluoride concentrations below 1 mg L^{-1} in drinking water.

No significant difference was found in fluoride levels between Iraqi bottled water and those imported from Iran, Turkey, Kuwait and Jordan, whereas there was a remarkable difference with all the 6 Saudi brands of bottled water which range from 0.793 to 1.052 mg L^{-1} (Table 2). The last difference support the assumption that Saudi packaged water may be fluoridated.

The fluoride levels in the present work are very close to those recorded in Turkey (Tokalioglu *et al*., 2004), Beljium (Vandevijvere *et al*., 2009), England (Zohouri *et al*., 2003), Australia (Cochrane *et al*., 2006),

Table 2: Fluoride content (mg L^{-1}), pH and EC (µS cm^{-1}) of imported bottled water samples occasionally consumed in Babil-Iraq

Water type	Source of water	Fluoride label information	Average fluoride concentration*	Conductivity	pH
Hana	Saudi Arabia	0.85	0.846	274	7.24
Aquafina	Saudi Arabia	1.00	1.052	213	7.00
Lama	Saudi Arabia	0.85	0.946	271	6.99
Sahatak	Saudi Arabia	0.80	0.793	167	7.01
Berain	Saudi Arabia	0.90	0.850	209	7.00
Hayat	Saudi Arabia	0.85	0.903	234	6.88
Aquafina	Kuwait	NL	0.031	219	6.69
Rawdatain	Kuwait	NL	0.093	304	7.84
Sama	Jordan	NL	0.180	642	7.68
Zamzam	Iran	NL	0.064	305	7.32
Taksu	Iran	NL	0.066	276	7.42
Hamidiya	Turkey	NL	0.029	68	7.04

NL: Not labeled; *All averages derived from five readings with a maximum standard deviation of 15%

USA-Ohio (Lalumandier and Ayers, 2000), Iran (Ghaderpoori *et al*., 2009; Amanlou *et al*., 2010; Dobaradaran *et al*., 2008), Brazil (De Souza *et al*., 2009), Greece (Ahiropoulos, 2006), Poland (Opydo-Szymaczek and Opydo, 2009) and Mexico (Jimenez-Farfan *et al*., 2011) which report average fluoride concentrations varying from 0.06 to <0.74 mg L^{-1}. The general range and pattern found in this study appears to be universal although the variation in mean values from study to study is significant. However, higher levels of fluoride (up to 4.8 mg L^{-1}) in bottled drinking water has also been reported (Ahiropoulos, 2006; Grec *et al*., 2008).

In comparison to the desirable and maximum permissible limit for fluoride in drinking water determined by WHO and Europeans' Directive (1.5 mg L^{-1}) or by Bureau of Iraqi Standards (1 mg L^{-1}) (WHO, 2006; Indermitte *et al*., 2009), the fluoride levels of the tap and bottled drinking water in Babil and may be in Iraq are rather low.

Fluoride intake: Total daily fluoride exposure can vary markedly from one region to another. This depends on the concentration of fluoride in drinking water and the amount of water drunk, levels in foodstuffs and the use of fluoridated dental products (WHO, 2008). Whilst almost all foodstuffs contain at least traces of fluoride, water and non-dairy beverages are the main sources of ingested fluoride, accounting for 66 to 80% of fluoride intake in US adults according to the concentration of fluoride in the public drinking water (WHO, 2005).

The average quantity of water consumed by Iraqi adults in Babil was estimated (via two questionnaires) to range from 800±240 mL in winter to 2000±650 mL in summer daily, compared with 2000 mL commonly used by WHO in computing drinking water guidelines and standards (WHO, 2005). Based on these volumes and the average fluoride levels presented in Table 1, the average daily intake of fluoride by Iraqi consumers from tap and bottled water are estimated to be 0.147±0.055 to 0.368±0.145 and 0.058±0.056 to 0.146±0.140 mg, respectively. These values matched well those recently reported by other workers for fluoride intakes in non-fluoridated communities assuming body weights of 70 kg for individuals (Health Canada, 2010).

Because data are not available to determine an Estimated Average Requirement (EAR), the reference value that was adopted in the present work is the American Canadian Adequate Intake (AI). The AI is based on estimated intakes that have been shown to reduce the occurrence of dental caries maximally in a population without causing unwanted side effects including moderate dental fluorosis. The adequate intake values are 3.8 mg day^{-1} for males and 3.1 mg day^{-1} for females assuming a reference weight of 76 and 61 kg for males and females respectively (Institute of Medicine IOM, 1997). These reference values are much higher than the fluoride intakes estimated for Iraqis, indicating that whether tap or bottled water are used as the primary source of drinking water, then Iraqi consumers are at a higher risk of dental caries.

Recent surveys of South Australian school children have provided strong evidence that the incidence of dental caries is increasing for the first time in decades. One of many possible contributing factors for this change in caries incidence may be the increased use of bottled water as a source of drinking and cooking water (Cochrane *et al*., 2006). These results confirm the need for a well organized water fluoridation program in Iraq.

Fluoridation: The results of the original epidemiological studies of the relationships between the concentration of fluoride in drinking water and dental caries indicated that reduction in the average number of dental caries per child was nearly maximal in communities having water fluoride concentrations close to 1.0 mg L^{-1}. This is how 1.0 mg L^{-1} became the "optimal" concentration (WHO, 2002) and artificial fluoridation of reticulated drinking water (elucidated over 60 years ago) has been instituted in many countries to reduce the incidence of dental caries.

Water fluoridation is found to be effective at reducing caries and has been hailed as one of the 10 greatest achievements in public health in the 20th century (Thippeswamy *et al*., 2010; Griffin *et al*., 2007; Centers for Disease Control and Prevention, 1999). The outcome of 120 fluoridation studies from different countries have shown ranges of reductions in dental caries of between 40-60% for permanent teeth and 50-60% for primary teeth in children aged 5 to 15 years who consumed fluoridated water for long periods (Zohouri *et al*., 2003). Reduction in dental caries has also been reported for all age groups up to 65 years when fluoridated water is consumed (Zohouri *et al*., 2003).

Worldwide around 355 million people are receiving artificially fluoridated water. In addition, around 50 million people receive water naturally fluoridated at a concentration of around 1 mg L^{-1} (WHO, 2005). Currently, countries like USA, Canada, Brazil, Argentina, Columbia, Chile, Australia, New Zealand, Malaysia and cities like Hong Kong and Singapore (Pizzo *et al*., 2007) all employ fluoridation schemes. The Australian Dental Association Policy Statement 2007 states that 'water fluoridation continues to be the most cost-effective, equitable and safe means to provide protection from dental caries and has been successfully utilized in Australia for over 50 years' (FSANZ, 2009).

In the light of these directives, it is recommended that drinking water in Iraq should be fluoridated. The optimal concentration of fluoride varies according to climatic conditions with the range 0.5-1.0 mg L^{-1} being generally recommended (WHO, 2005).

Optimizing fluoride levels in water supplies is an ideal public health measure because it is effective and inexpensive and does not require alert daily cooperation from individuals. Fluorine can be added to water by either adding a slurry of sodium fluorosilicate, a solution of hydrofluorosilicic acid or less commonly a saturated solution of sodium fluoride. These solutions are added using a metering dosing solution to ensure accurate addition of fluoride into the water flow.

Labeling of bottled water: Regarding the quality of the labeling of bottled water, 19 of the Iraqi water samples (63.5%) did not state the fluoride concentration on the label (Table 1), while the labeled bottles did not display precise values for fluoride content. One of the samples (Rovian) listed the level at <0.11 mg L^{-1} fluoride on the label compared with a measured fluoride content of 0.242 mg L^{-1}. Another brand (Nabaa Alfurat) was found to contain 0.056 mg L^{-1} fluoride but it listed a fluoride level at 0.7 mg L^{-1} on the label. Within the imported samples only Saudi bottled water stated the fluoride level and presented reliable agreement between the fluoride content measured and that shown on the product label. These discrepancies were commonly found even in developed countries (Zohouri *et al*., 2003).

So far no strict regulation on the labeling of fluoride contents of bottled drinking water in Iraq; thus, appropriate regulation seems to be necessary. Strategies such as direct periodic inspection of the producing plants and application of warning and penalization measures may cause regularization of the fluoride levels found in bottled water. The inclusion of the actual fluoride content on the label would allow the consumer to be aware of the presence or absence of fluoride in drinking water. This way, the consumer would be able to know the amount of ingested fluoride and then make an informed decision about the choice of drinking water.

CONCLUSION

The tap and bottled water are the main sources of drinking water for people of Babil, Iraq. After evaluating the fluoride concentration by ionic selective electrode, it is found that the level of fluoride is far below the upper level recommended by WHO and by Bureau of Iraqi Standards. To prevent dental caries, it is recommended that drinking water in Iraq should be fluoridated. Regarding the quality of the labeling of bottled water, most of the Iraqi water samples did not state the fluoride concentration on the label, while the labeled bottles did not display precise values for fluoride content. Strategies such as direct periodic inspection of the producing plants and application of warning and penalization measures may cause regularization of the fluoride levels found in bottled water.

The results of the present study as well as other available data from water quality should be taken into account when developing strategies for safe drinking water supplies.

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