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Fluoride Content of Still Bottled Water Brands Available in Enugu, Nigeria

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Abstract

Fluoride content of drinking water is important for public health because of its ability to prevent dental cavities in optimal dose. Twenty different brands of still bottled water brands were purchased from supermarkets in Enugu metropolis from October 2017 to March 2018. Three bottles of different batch numbers from each of the brands were used for the analysis. The fluoride content of all samples was determined, in triplicate per batch, using the zirconium- Alizarin red-S spectrometric method. The mean (± SD) fluoride content of the bottled waters was 1.66 (± 0.28) mg L-1 with a range from 1.35 - 2.25 mg L-1. One-way ANOVA showed a significant difference in the fluoride content of the batches, *p* - value, 0.008<0.05. The paired samples t-test showed a non-significant difference between the first and second reading of each batch, *p*-value, 0.761>0.05. The independent samples t-test showed a non-significant difference between mean fluoride content of bottled water made in Enugu and those made outside Enugu, *p*-value, 0.290>0.05. The result from this study showed that different brands of bottled water consumed in Enugu metropolis contain appreciable fluoride contents which could help to prevent dental problem in the consumers.

Key words: fluoride content, bottled drinking water, Alizarin red spectrometric analysis, samples

INTRODUCTION

Fluoride is a solid naturally occurring chemical compound that is found in all natural water sources (McGRADY *et al.*, 2010). It is also the ionic compound of the trace element fluorine, which is commonly found in the surroundings. As a water-soluble mineral, fluorine is drained by rain water from soil and rocks into water found underground in the cracks and spaces in soil, sand and rock (McGRADY *et al.*, 2010). Fluoride is a harmless and result-oriented material that can be used to stop and remediate tooth decay when used as prescribed or consumed in fluoride containing water (CDC 2018, 2001). The formation of tooth decay depends on a lot of factors and over time, can climax in concentrated breakdown of hard dental tissues (enamel, dentin and cementum) by the weak acids produced by naturally occurring oral bacteria when they breakdown food debris or sugar on the tooth surface. (CDC, 2001, SELWITZ *et al.*, 2007 and ADAIR, 2006). Acids leach minerals from the enamel through a process called demineralization. Fluoride replaces the lost calcium hydroxyapatite structure in the enamel leached by these acids by forming calcium fluorapatite, which is more resistant to acid attacks (McGRADY *et al.*, 2010). The replacement of these acid-leached minerals by fluoride can both reverse the early decay process as well as create a tooth surface that is more resistant to decay (ADAIR, 2006). The sources of human exposure to systemic and topical fluoride include drinking fluoridated water, beverages and food prepared with fluoridated water (ABOULEISH, 2012 and JAGTAP *et al* 2012), fluoridated salt and milk (WHELTON *et al.*, 2004) oral prophylactic procedures (RAJKOVIC and NOVAKOVIC, 2007) air (JAGTAP *et al.*, 2012), fluoridated gel dentifrices (CDC, 2013), oral rinsing solutions and cosmetics (JAGTAP *et al* 2012), dietary fluoride supplements (drops, tablets and lozenges) (JAGTAP *et al.*, 2012). Drinking water is the major route of daily oral intake of fluoride (MURRAY, 2003). For mature people, the consumption of food and ingestion of drinking-water are the main paths for the intake of fluoride (W.H.O, 2011 and WIKIPEDIA, 2018). The recommended level of fluoride for optimal dental health, is currently set at 0.7 parts fluoride per million parts water (ppm) (H.H.S., 2017).

Through the process of water fluoridation, the fluoride content of fluoride-deficient water is adjusted to this recommended level (H.H.S. 2017 and U.S P.H.S., 2015). Intake of fluoride-deficient water may result in tooth decay and dental caries (FAWELL *et al* 2006). On the other hand, high fluoride content in water, may cause dental and skeletal fluorosis (FAWELL *et al.*, 2006). The severity of occurrence depends upon the dose and the period of time of intake. Dental fluorosis occurs when consumption of high doses of fluoride results in teeth striation. Thus the appearance of the dentition becomes aesthetically objectionable. Infants under the age of 6 are more vulnerable to dental fluorosis. This is due to the fact that the enamel formation is not yet complete. Dental fluorosis happens more often where teeth are breaking out under the gums. Skeletal fluorosis is a bone disease solely caused by excessive accumulation of fluoride in the bones and in advanced cases causes painful damage to bones and joints (FAWELL *et al.*, 2006). This becomes more obvious as one grows older with ingestion of high levels of fluoride. The maximum allowable concentrations of fluoride in drinking water is shown in Table 1. These guidelines are designed to prevent the public from being exposed to harmful levels of fluoride. (H.H.S 2003, W.H.O., 2011).

Table 1: Fluoride Guideline Standard for Drinking Water.

State/Organization	Recommended Min. Value (mg/L)	Max. Value (mg/L)	Reference
WHO	0.5	1.5	WHO [2011]
USA – Primary	0.5	4.0	
Secondary	0.5	2.0	USEPA [2002]
Nigeria*	0.5	1.5	Standards Organization of Nigeria : NIS 554 (2007)

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Source: (U.S.E.P.A., 2002 and S.O.N. 2007).

Though the oral health of the public is depreciating, Nigeria has no clear legal framework for water fluoridation despite having an oral health policy (OLUSILE, 2010). Water fluoridation from the public water mains would only affect a few people since the taps are drying up in many cities and States. An assessment in Nigeria shows that about 21% of natural water sources have a fluoride range of 0.3-0.6 ppm, while more than 17% of water sources elsewhere have fluoride content above this range (AKPATA *et al.,* 2009). Many rural and urban households in Nigeria, have limited access to safe drinking water. This limited access to safe drinking water, the hot weather, coupled with population and urbanization has fueled the sale of bottled water (EUROMONITOR, 2018). Bottled water has been described as water that is intended for human use and closed up securely in bottles or other containers with no added constituents, except that it may contain a harmless and appropriate agent that stops the growth of microorganisms (FDA WEBSITE). There is probable apprehension in Nigeria as well as other countries about the purity and the presence of contaminants in natural water supplies which has contributed to the replacement of peoples' daily water intake by bottled water (AHIROPOULOS, 2006). Despite the large demand for bottled water in Nigeria, the water quality of commercially bottled water is unclear. Makers of bottled water brands sometimes list the nutritional content of their products but labeling of the fluoride content of the products is not legally required in Nigeria.

The aims of this study therefore are to assess fluoride concentration in bottled water and confirm the accuracy of the labelling of fluoride levels in bottled water. This study is meant to create awareness on the status of the fluoride content and hence the potential or otherwise for causing dental cavity, dental fluorosis or skeletal fluorosis of these bottled water brands in consumers.

MATERIALS AND METHOD

2.1. Bottled Water Samples:

Twenty brands of bottled water where obtained from supermarkets in Enugu, Nigeria. 9 brands where produced in Enugu while 11 brands where produced outside Enugu. Three bottles from each brand, each with a different batch number and date of bottling were purchased over a period of six months. All bottles were stored in a dark place and in their original closed containers at room temperature until the fluoride analysis was made. The bottled water containers were made of plastic and the volume varied between 500 – 1500 ml.

2.2. Fluoride Determination:

Fluoride ion content in the bottled water brands was determined by the Zirconium-Alizarin red-S spectrophotometric method (APHA, 1998). In this technique zirconium oxychloride and a derivative of a dye named alizarin red (1,2-dihydroxyanthraquinone) produce a red complex. Fluoride ion, depending on its concentration bleaches this complex from red to yellow until it forms a colourless solution. Variations in the colour of the solution with regards to standard fluoride ion solution, were determined with a UV-visible spectrophotometer. 0.75 g alizarin red indicator was dissolved in distilled water in a 1,000 ml volumetric flask. Also, 0.345 g of zirconyl chloride was dissolved in distilled water and then 33.3 ml and 101 ml of concentrated sulfuric acid and hydrochloric acid were added to the solution respectively. The final volume of the solution was made up to 1,000 ml by adding distilled water. 5 ml of alizarin red and zirconyl chloride-acid solutions were added to 100 ml of each batch of the water brands and the standard solutions respectively. They are allowed to stand for 60 minutes for complete colour development. The absorbance of the solutions was thereafter read at 520 nm with a spectrophotometer (GE Amersham Pharmacia Biochrom Ultrospec 2100 ***Pro***). The readings of each batch was done in triplicate then the average was recorded. Fluoride content was determined by comparing the light absorbance of each solution to the standard fluoride solution (NH4F.HF dissolved in distilled water) using a calibration curve. To assess the reliability of the method, one batch number (of three) for each of the bottled water samples was randomly selected and the samples were re-analyzed. pH values of the samples were determined using METTLER TOLEDO Seven Compact pH/Ion meter and Inlab Expert Pro-ISM pH electrode.

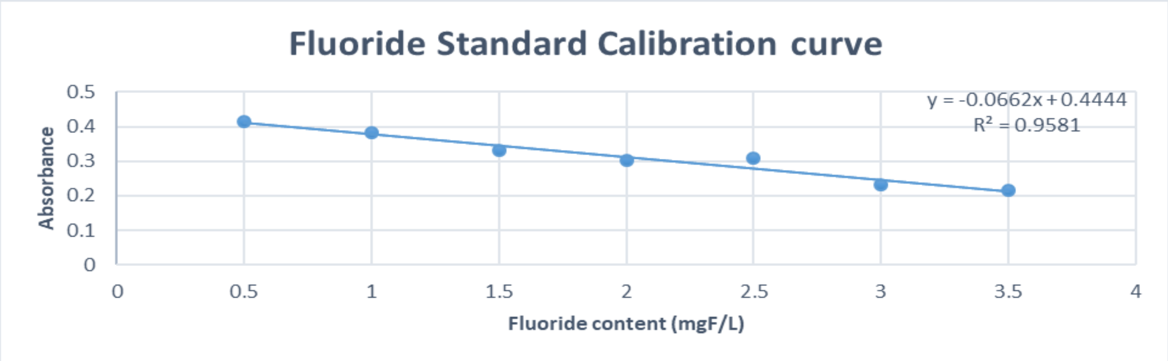

Fig. 1: The calibration curve for determining fluoride content in bottled drinking waters.

2.3 Data Management:

Final calculation of the fluoride content of the samples from the absorbance readings was carried out in Microsoft EXCEL 2016 using linear regression. IBM-SPSS Statistics 23 software was used for the statistical evaluation of the data obtained and presented as mean ± standard deviations. The paired samples t-test was employed to compare the first and second measurements of each batch. The one-way ANOVA test was used to compare the fluoride content among the different batches of the same brand and the Independent samples t-test was used to evaluate the differences between water bottled in Enugu and those bottled in other parts of Nigeria. Differences between means were considered to be significant when $p < 0.05$.

3. Results:

The linear calibration curve was obtained by plotting absorbance readings versus mg/L fluoride content in bottled drinking waters (Figure 1). The fluoride calibration curve gave the following linear regression model:

$$y = -0.662x + 0.4444 \ (R^2 = 0.9581) \qquad \text{(Eq. 1).}$$

The fluoride content shown in mg Fluoride/L and the pH values of the 20 different types of bottled water are shown in Table 2. The mean (±SD) fluoride content for all brands sampled was 1.66 (±0.28) mg F/l, ranging from 1.35 - 2.25 mg F/l. The pH values of the samples ranged from 4.91-7.99.

The accuracy of the method for fluoride determination was confirmed by comparing the first and second readings of each batch using the paired sample t-test which showed that there were no statistically significant variations between these readings, $p = 0.761$. The highest mean fluoride concentration by brands was found in Swan water from Kerang, Mangu, Plateau State, Northcentral Nigeria, [2.25 (±0.16) mg F/l]. All the brands did not list their fluoride content on their labels except Ragolis water. Between subjects and group ANOVA statistics indicated significant variations in fluoride content among the three batches for each brand, $p = 0.008$. The variations in the fluoride content of samples produced in Enugu were not significant, $p = 0.079$ while the variations in the fluoride content of samples produced outside Enugu were significant, $p = 0.023$. The Independent t-test conducted showed that the mean fluoride content of samples produced outside Enugu [1.70± (0.28) mg F/l] was not significantly higher than the mean fluoride content of samples produced in Enugu [1.62± (0.28) mg F/l], $p = 0.290$.

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Table 2: Fluoride content of commercially available bottled water in Enugu.

Bottled water brand	Source	Fluoride content (mg F/l)			Mean (±SD) (mg F/l)	Labeled content (mg F/l)[a]	pH of sample
		Batch 1	Batch 2	Batch 3			
Swan	Kerang	2.41	2.24	2.09	2.25±0.16	NM[b]	7.37
Bigi	Ikogosi	1.30	2.08	1.50	1.62±0.39	NM	6.15
Ragolis	Lagos	1.22	1.52	1.41	1.38±0.15	0.05	6.92
Lucozade Hydropure	Ogun	1.44	1.82	1.56	1.61±0.19	NM	7.99
Eva	Lagos	1.81	1.55	1.08	1.48±0.37	NM	7.09
Aquafina	Lagos	1.41	1.79	1.97	1.72±0.29	NM	6.13
Nestle’	Lagos	1.62	1.70	1.77	1.70±0.08	NM	6.43
La Sien	Port Harcourt	2.02	1.73	1.55	1.76±0.24	NM	7.89
Gossy	Ikogosi	1.80	1.70	1.77	1.76±0.05	NM	5.34
Event	Anambra	1.68	1.75	1.65	1.70±0.05	NM	4.91
Cascade	Lagos	1.56	1.64	1.94	1.71±0.20	NM	6.45
Aquarapha	Enugu	1.43	1.79	1.79	1.67±0.21	NM	6.64
Earnwell	Enugu	2.09	2.09	2.03	2.07±0.04	NM	7.06
Bejoy	Enugu	1.25	0.96	1.85	1.35±0.45	NM	5.94
Ivy	Enugu	1.40	1.40	1.40	1.40±0.00	NM	6.87
Rovia	Enugu	1.80	1.88	1.26	1.65±0.34	NM	7.29
Trinity	Enugu	1.74	1.62	1.58	1.67±0.06	NM	5.93
Exalte’	Enugu	1.65	1.64	1.61	1.63±0.02	NM	5.90
Ezbon	Enugu	1.59	1.70	1.41	1.57±0.15	NM	5.62
Jasmine	Enugu	1.88	1.64	1.18	1.56±0.36	NM	6.86

[a]mg/L fluoride is equivalent to ppm fluoride [b]Not mentioned

Discussion:

The pH values obtained showed that 10 table water brands had pH values within the W.H.O acceptable limits (6.5 - 8.5), while the rest of the water brands fell below the permissible limits. Water pH is classified as a secondary drinking water contaminant whose impact is considered aesthetic. Water with low pH value can be acidic, naturally soft and corrosive. It can also cause gastro-intestinal irritation in sensitive individuals (WHITE, 1985). The Nigeria drinking water standard for fluoride in tap and bottled water according to Standards Organization of Nigeria, S.O.N is the same as the W.H.O drinking water standard which is 1.5 mg/L (U.S.E.P.A., 2002, S.O.N., 2007 and W.H.O, 2011). However, the United States Food and Drug Administration, FDA standard of quality (SOQ) states that domestic bottled water with no added fluoride may contain between 1.4 and 2.4 milligrams fluoride per litre, depending on the annual average of maximum daily air temperatures at the location where the bottled water is sold at retail (CDC., 2013). Four bottled water brands namely Ragolis, Eva, Bejoy and Ivy at 1.38, 1.48, 1.35 and 1.40 mg F/l respectively had fluoride levels below the S.O.N and W.H.O permissible limit value of 1.5 mg F/L. The rest had fluoride content above this value. Compared, however to FDA standard of quality, all the water samples are within the standard limit range of 1.4 - 2.4 mg F/L The variation of naturally occurring fluoride in groundwater which is the major source of water for the water bottlers in Nigeria depends on the type of aquifer. These values vary from 1 - 8 mg F/L for crystalline basement aquifers (LAR *et al.*, 2007) and 1 - 4 mg F/L for sedimentary aquifers (DIBAL and LAR, 2005). The U.S. Department of Health and Human Services (HHS) recommended an optimal fluoride level of 0.7 mg/L fluoride (FDA WEBSITE). Comparing the fluoride levels in the bottled water brands analyzed in this work with the HHS recommended optimal value of 0.7 mg F/L shows that all the samples have fluoride levels higher than this value. This implies that the fluoride levels in these samples may either promote healthy teeth or cause dental and skeletal fluorosis depending on the dose consumed. In this study, the highest mean fluoride content of bottled water was 2.25 mg F/L. Higher fluoride content of up to 4.14 mg F/L (MOHAMED, 2016) and 4.8 mg/L (AHIROPOULOS, 2006) have been reported in bottled water. In other studies, also lower fluoride levels in bottled water have been determined: 0.04 - 0.3 mg F/L (TARUN *et al.,* 2017) and 0.1 - 0.8 mg F/L (TOUMBA *et al.,* 1994). The value of 2.25 mg F/L of Swan water is higher than 0.14 - 0.41 mg F/L reported for spring water in Kerang province, Plateau State ((DIBAL and LAR, 2005). This increase may be attributable to seasonal variation or artificial fluoride may have been added to boost the fluoride content to optimal level.

Conclusion:

The bottled drinking water brands tested contain differing fluoride content. Though the mean content of fluoride in the water brands are above W.H.O. and the Nigerian limit value, they are within the standard limit range set by U.S. FDA. The fluoride levels in these brands can therefore prevent dental cavities, but their potential in causing dental or skeletal fluorosis depends on the amount ingested over a time period. Labelling of fluoride content in table water would enable the populace make informed choices about the level of their exposure to fluoride.

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