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FLUORIDE IN BLACK AND GREEN TEA (*CAMELLIA SINENSIS*) INFUSIONS IN ETHIOPIA: MEASUREMENT AND SAFETY EVALUATION

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ABSTRACT. The fluoride contents in the infusions of 21 commercially available Ethiopian and imported black and green tea brands; in leaf and bag forms was determined by a fluoride ion-selective electrode method. Of the samples analyzed twelve were products from Ethiopia and the remaining nine were imported tea brands. The effect of brewing time on fluoride release from tea was assessed. Results showed that fluoride release increased with increasing brewing time (3, 5 and 10 min). Fluoride level after 5 min brewing for black tea leaves, green tea bags, and black tea bags was in the range of 117–682 mg/kg, 111–190 mg/kg and 141–246 mg/kg, respectively. The WHO guideline for daily fluoride intake is 2 mg for children and 4 mg for adults. Assuming that one consumes 4 cups of tea everyday (400 mL) and each cup uses 2.5 g of tea leaves, the daily fluoride intake from black tea leaves may be in the range between 1.11 and 6.82 mg. For the same condition, if consumption of one green tea bag is considered, the fluoride intake can be in the range between 1.00 and 1.38 mg. Similarly, intake from the black tea bags may range from 0.86 to 1.81 mg. Considering the Ethiopian black tea alone, the daily fluoride intake may range from 2.48 to 6.82 mg. Thus according to the WHO recommendation for daily fluoride intake and ignoring other possible sources; the black and green tea bags and imported black tea leaves are safe for all age groups. None of the 10 Ethiopian black tea leaf brands are safe for children but 30% of the analyzed samples are safe for adults.

KEY WORDS: Black tea infusion, Green tea infusion, Fluoride intake, Brewing time, Safety evaluation

INTRODUCTION

Fluoride (F^-) is an important anion, present in various environmental, clinical and food samples. Small amounts of fluoride are vital for human in preventing dental carries, but it is toxic in larger amounts. For adults the lethal dose is 0.20–0.35 g F^- per kg body weight [1-3]. Fluoride is widely used in various branches of industry and some fluoride compounds are formed as by-products in certain processes. Excessive amounts of fluoride in the form of different compounds can enter the human body by means of polluted air, water and the food chain. An additional source of fluoride for humans is toothpastes containing 0.1% fluoride in the form of NaF, SnF_2, Na_2PO_3F and water fluoridation. Fluoride has also been used to treat osteoporosis [1, 2]. It is very characteristic that fluoride prevents tooth decay at about 1 mg/L but causes mottled teeth (dental fluorosis) and bone damage (skeletal fluorosis) at around 5 mg/L over prolonged periods of exposure. In dental fluorosis the structural integrity of the enamel is affected and small pits are left in teeth as it breaks away. While skeletal fluorosis is the accumulation of fluoride in skeletal tissues associated with pathological bone formation [4-7].

Rocks, soil, water, air, plants, animals and foodstuffs all contain fluoride in widely varied concentrations [8]. Tea plant (*Camellia sinensis*) accumulates and stores more fluoride by absorbing it from the air and soil than any other plant. Up to 98% of the fluoride in tea plants is stored in leaves [9, 10]. Thus, tea leaves are usually very rich in fluoride. The fluoride content increases with the maturation stage of the leaves.

Fluoride content in tea has increased dramatically over the past 20 years due to industrial contamination [11]. Fluoride content of 100–430 mg/kg can be found in the delicate buds and young leaves, which are usually made into green tea or black tea, while fluoride levels of 530–2350 mg/kg can be found in the oldest leaves, which are not used as tea [9, 10, 12]. A

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substantial amount of fluoride is released during tea infusion and about 95% of the released fluoride is available to consumers [13].

During recent years several papers have been published on the fluoride content of tea infusions of different origin and type using ISE method [9-15]. Fluoride level of 352–576 mg/kg and even as high as 1175 mg/kg in Chinese brick tea [14], 2.10–123 mg/kg in Chinese green and black teas, 35–182 mg/kg in Iranian black teas [13], 97–148 mg/kg in black tea leaves and 139–223 mg/kg in black tea bags originated from United Kingdom, India, China, Japan, and Sri Lanka, and fluoride level of 311–604 mg/kg in some of black tea brands marketed in United States, United Kingdom, China and Hong Kong have been reported [9, 10].

Several papers have also been published on the fluoride content of tea leaves. However, the results obtained are often in poor agreement [15, 16]. The variation can be explained on the basis of the leaf age, maturity and genetics of the plant, rainfall, altitude, fertilizer, type of soil and water used for irrigation [16, 17].

In another study by Sukru *et al.* [18], extraction of fluoride from tea leaves to tea liquors was high with soft water than hard water. They have also seen the fluoride levels in tea liquors increases with the increase of brewing time. Cao *et al.* [11] has seen the effect of the paper bag used to store the black tea leaves and addition of milk and sugar supplements [19] on the ionic fluoride level in the tea filtrate. Their results reveal that neither addition of milk and sugar nor the paper bag has effect on the tea filtrate ionic fluoride level. The effect of acid on fluoride in the tea extract was also assessed by Sukru *et al.* [18], which showed that the fluoride concentrations in the tea infusions were not changed by the addition of 1 M perchloric acid. Therefore, addition of lemon or lemon juice, which is a common way of drinking tea in some countries including Ethiopia, is not expected to affect the quantity of free fluoride in tea.

Natural fluoride from drinking water and food, especially tea infusions and high fluoride salts are the main sources of the total fluoride intake of the population [20]. In Ethiopia the concentration of fluoride in surface water is generally below 1.5 mg/L in highland areas except in some locations whereas in lowlands mainly in the Rift Valley Regions, the fluoride content of drinking water from ground water is as high as 33 mg/L. Fluorosis resulting from the intake of tea was reported in different parts of the world [9, 10, 12, 14, 20].

Kloos and Redda [21] reported that water as epidemiologically the most important source of fluoride (75–90% of daily intake) in most areas of the Rift Valley of Ethiopia. Others indicate that considerable exposure risk is also associated with the consumption of fish bones, canned meat, vegetables, grains and other staples, local salt, drinks (especially tea) and air [22-26]. In some African and Asian communities, intake of fluoride from food has been found to be higher than from water [27]. Tea is considered to be one of the major sources of fluoride next to water.

Tea infusions are the most popular beverage consumed by human society worldwide, second only to water [28]. Tea consumption may also provide an appreciable proportion of the daily dietary intake of fluoride [29-31].

The quality of infused tea depends on the percent of extraction, which in turn is a function of type, strength, and duration of infusion. Boiling increases the fluoride extraction, but also affects the flavor of tea. Five minutes produces the best flavor with least extraction of unpleasant tasting tannin [16].

The objective of this study was to determine the fluoride level of tea infusions prepared from commercially available Ethiopian and imported black and green teas and to evaluate safety levels.

EXPERIMENTAL

Sampling

A total of 21 tea brands, which are commonly available in the Ethiopian markets were collected from different shops and supermarkets in Addis Ababa. Of the total 21 tea brands, three (14%) were green tea and the rest 18 (86%) were black tea products. Three packets of twelve brands tea and two packets of nine brands tea were bought from either one shop or from different shops depending upon the availability of the particular tea brand in different shops for the reason of greater homogeneity. Detailed description of the tea brands and the amount of samples collected from each brand are summarized in Table 1.

Table 1. List of sampled tea.

No.	Tea brand name	Tea form	Amount per box	Country of origin	Country packed in	Area grown	Sample collected, box
Green tea bags (GTB)							
1	Ethiopian green tea	Leaf	25 bags	Ethiopia	Ethiopia	W & GTP	3
2	Hyson green tea*	Leaf	25 bags	Sri Lanka	Sri Lanka	NA	2
3	Quality green tea*	Leaf	25 bags	Sri Lanka	Sri Lanka	NA	2
Black tea leaves (BTL)							
4	Abay	Leaf	100 g	Ethiopia	Ethiopia	W & GTP	3
5	Abyssinia	Leaf	100 g	Ethiopia	Ethiopia	W & GTP	3
6	Addis	Granular	100 g	Ethiopia	Ethiopia	W & GTP	3
7	Ahadu	Leaf	100 g	Ethiopia	Ethiopia	W & GTP	3
8	Almeta	Leaf	100 g	Ethiopia and Kenya	Ethiopia	K,W & GTP	3
9	Anbessa	Leaf	100 g	Ethiopia	Ethiopia	CTP	3
10	Eirmon	Leaf	100 g	Ethiopia	Ethiopia	W & GTP	3
11	Gumaro	Leaf	100 g	Ethiopia	Ethiopia	GTP	3
12	Haron	Leaf	100 g	Ethiopia	Ethiopia	W & GTP	3
13	Hyson*	Leaf	200 g	Sri Lanka	Sri Lanka	NA	2
14	Lipton*	Leaf	250 g	China	Saudi Arabia	NA	2
15	Mohmood*	Granular	200 g	Kenya, India and Sri Lanka	Sri Lanka	NA	2
16	Wushwush	Leaf	100 g	Ethiopia	Ethiopia	WTP	3
17	Quality*	Leaf	150 g	Sri Lanka	Sri Lanka	NA	2
Black tea bags (BTB)							
18	Addis	Leaf	25 bags	Ethiopia	Ethiopia	W & GTP	3
19	Hyson*	Leaf	25 bags	Sri Lanka	Sri Lanka	NA	2
20	Lipton*	Leaf	25 bags	China	UK	NA	2
21	Lipton flavored black tea*	Leaf	25 bags	China	UK	NA	2

*Imported ones, W & GTP = Wushwush and Gumaro tea plantations, NA = not available, K = Kenya, CTP = Chewaka tea plantation, GTP = Gumaro tea plantation, WTP = Wushwush tea plantation.

Preparation of tea infusion samples

Infusions of stick and granular tea samples were prepared by employing customary way of tea preparation. In this procedure, distilled-deionized water was boiled in 100 mL beaker; 2.5 g of tea leaves was added to the beaker and allowed to infuse for 3 min. The tea was filtered using a plastic filter (mesh) and allowed to cool to room temperature. The infusion was filtered again using Whatman No. 42 filter paper. Finally the volume of the infusion was made 100 mL with distilled-deionized water again to compensate for the loss during boiling prior to storage in

plastic bottles. With similar procedure, tea infusions were prepared by brewing for 5 and 10 min in triplicate for all tea brands (i.e. a total of six or nine infusions were prepared for each tea brand).

Preparation of infusions from tea bag samples involved immersion of the bags in to distilled-deionized water which was boiled in 100 mL beaker. Randomly selected 3 different tea bags of the same brand were put in each of the three beakers and infused for 3 min. The tea infusion was allowed to cool to room temperature. The infusions were filtered using a Whatman No. 42 filter paper. Volume of the infusions was made 100 mL again with distilled-deionized water. The infusions were stored in 3 separate plastic bottles. Similarly, tea infusions with 5 and 10 min brewing time were prepared in triplicate for all the tea brands.

Two or three packets of each tea brand were bought from either one shop or from different shops depending upon the availability of the particular tea brand in different shops for the reason of greater homogeneity. Three tea infusions were prepared from each of the two or three packets of the particular tea brand and triplicate measurements were made on each infusion. The results are reported as mean value of the 18 or 27 measurements [three measurements of 2 (packets) x 3 (infusions) or 3 (packets) x 3 (infusions) for each particular tea brand]. Similarly the precision of the measurements was expressed as standard deviation and relative standard deviation of the 18 or 27 measurements (three measurements of 2 x 3 or 3 x 3 infusions) for each particular tea brand.

Determination of fluoride

The fluoride content of the tea infusions was measured using pH/ISE meter (Orion model, EA 940 Expandable Ion Analyzer, USA) equipped with combination fluoride selective electrode (Orion Model 96-09, USA).

A 1000 mg/L fluoride stock solution was prepared by dissolving 2.21 g of anhydrous sodium fluoride (99.0% NaF, BDH Chemicals, England) in 1000 mL of distilled-deionized water in volumetric flask. By subsequent dilution of the stock solution 100, 20, 10, 5, 1, and 0.5 mg/L of F^- standards were prepared.

The total ionic strength adjustment buffer (TISAB) was prepared by mixing 57 mL of glacial acetic acid, 58 g of sodium chloride, 7 g of sodium citrate and 2 g of EDTA in distilled-deionized water in a 1000-mL volumetric flask, its pH was adjusted to 5.3 with 6 M sodium hydroxide and diluted to the mark with distilled-deionized water. A five points calibration curve was prepared using 10 mL of the 20, 10, 5, 1, and 0.5 mg F^-/L standard solutions together with 10 mL of the TISAB.

Evaluation of analytical methods

In order to investigate the accuracy and precision of the method, three tea samples were brewed using distilled-deionized water spiked with a fluoride standard solution and analyzed by the described method. The accuracy expressed as percent recovery was obtained by comparing the results between the fluoride found and the fluoride added where as the precision of the method was determined as relative standard deviation. Table 2 presents the accuracy and precision of the method. The method detection limit which is defined as the least concentration of the analyte that can be detected within a predetermined level of confidence and is equal to three times the standard deviation of the blank [32] was also determined by taking ten blank solutions. As a result the method detection limit found was 0.035 mg/L which is lower than the instrument detection limit 0.02 mg/L.

Table 2. The accuracy and precision of the method.

Tea brand	F^- added, mg/kg	Amount recovered, mg/kg	Recovery (%)	Precision (% RSD)
Ethiopian GTB	40	37.4 ± 2.9	93.4 ± 7.3	7.8
Addis BTP	80	70.7 ± 3.7	88.4 ± 4.6	5.2
Lipton flavored BTB	40	38.5 ± 2.4	96.1 ± 5.9	6.2

RESULTS AND DISCUSSION

Level of fluoride in tea infusions

The fluoride concentrations (mg/kg) of the infusions prepared after 3, 5 and 10 min brewing time are shown in Tables 3-5. The fluoride concentration of the tea infusions was in the range of 80–634 mg/kg for 3 min brewing time, 111–682 mg/kg for 5 min brewing time, and from 130–728 mg/kg after 10 min brewing time. The fluoride content of green tea bags was in the range of 80–158 mg/kg for 3 min infusion, 111–190 mg/kg for 5 min infusion, and 130–245 mg/kg for 10 min brewing time. For the black tea leaves analyzed the fluoride concentration was in range of 82–634 mg/kg, 117–682 mg/kg, and 153–728 mg/kg for 3, 5, and 10 min brewing time, respectively. Black tea bags fluoride content was found to be in the range of 110–189 mg/kg, 141–246 mg/kg, and 167–298 mg/kg for 3, 5, and 10 min brewing time, respectively.

Table 3. Range of fluoride levels of tea infusions for 3 min brewing time for 2 x 3 or 3 x 3 samples of each tea brand taken from one or different shops (each sample was measured three times).

Tea brands name	Range, mg F^-/kg	[*]Mean F^-, mg/kg	% RSD
Green tea bags			
Ethiopian green tea	145 – 147	146 ± 1	0.6
Hyson green tea	78 – 80	80 ± 1	1.4
Quality green tea	155 – 159	158 ± 2	1.4
Black tea leaves			
Abay	632 – 637	634 ± 3	0.4
Abyssinia	547 – 554	551 ± 3	0.6
Addis	290 – 292	291 ± 1	0.4
Ahadu	546 – 550	548 ± 2	0.3
Almeta	192 – 193	192 ± 1	0.2
Anbessa	398 – 400	399 ± 1	0.2
Eirmon	355 – 359	357 ± 2	0.6
Gumaro	340 – 347	343 ± 4	1.0
Haron	348 – 352	350 ± 2	0.7
Hyson	80 – 85	82 ± 3	3.0
Lipton	88 – 93	91 ± 2	2.6
Mohmood	118 – 119	118 ± 1	0.4
Wushwush	518 – 524	521 ± 3	0.7
Quality	178 – 183	180 ± 3	1.4
Black tea bags			
Addis	184 – 191	189 ± 4	2.1
Hyson	123 – 127	125 ± 3	2.0
Lipton	108 – 113	110 ± 2	2.0
Lipton flavored black tea	140 – 146	143 ± 3	2.0

[*]Values are mean ± SD (mg/kg) for n = 3, the mass of tea leaves in tea bags was calculated as mean of five tea bags.

The highest level of fluoride was found in the infusion of Abay black tea leaves with mean fluoride concentration of 634, 682, and 728 mg/kg for 3, 5, and 10 min brewing time, respectively. Whereas the lowest fluoride concentration was found in the infusion prepared from Hyson green tea bag, which is imported from Sri Lanka with mean fluoride level of 80, 111, and 130 mg/kg for 3, 5, and 10 min brewing time, respectively.

Comparison of fluoride concentration among the different tea brand infusions

The fluoride concentration of tea infusions varies considerably from one brand to the other, this might be due to the differences in the tea plant type, area of cultivation of the plant which may include soil type, altitude, quality of water used for irrigation, etc., harvesting time and harvesting system, i.e. type of leaf harvested. It was earlier shown that the variation is not due to the differences that exist in processing [18, 33-35].

For 3 min brewing time, except the Ethiopian green tea which is exceeded only by two imported tea brands; the top 11 with high level of fluoride are all produced in Ethiopia (Table 3). This high fluoride content of the Ethiopian teas may be due to the high soil fluoride level, low soil pH and calcium level which is used to precipitate fluoride in the soil and roots of the plant. But most probably it may be due to the usage of the cheap and highly profitable steam parts, older and low grade dust tea leaves together with the younger shuts of the plant. The older and low grade dust tea leaves are expected to have higher level of fluoride than the young shoots, the top 11 high-level infused fluoride are from the Ethiopian products [14, 15, 18, 33, 34].

Table 4. Fluoride level of the tea infusions for 5 min brewing time for 2 x 3 or 3 x 3 samples of each tea brand taken from one or different shops (each sample was measured three times).

Tea brands name	Range, mg F/kg	*Mean F, mg/kg	% RSD
Green tea bags			
Ethiopian green tea	159 – 161	160 ± 1	0.6
Hyson green tea	111 – 112	111 ± 1	0.8
Quality green tea	185 – 194	190 ± 5	2.4
Black tea leaves			
Abay	678 – 688	682 ± 5	0.7
Abyssinia	559 – 563	562 ± 3	0.5
Addis	306 – 311	308 ± 3	0.9
Ahadu	573 – 580	577 ± 4	0.6
Almeta	246 – 250	248 ± 2	0.9
Anbessa	441 – 445	444 ± 3	0.6
Eirmon	421 – 427	425 ± 3	0.7
Gumaro	400 – 407	403 ± 3	0.9
Haron	360 – 361	361 ± 1	0.2
Hyson	115 – 118	117 ± 1	1.2
Lipton	116 – 120	118 ± 2	1.7
Mohmood	138 – 140	140 ± 2	1.3
Wushwush	570 – 572	571 ± 1	0.2
Quality	193 – 195	194 ± 1	0.4
Black tea bags			
Addis	243 – 248	246 ± 3	1.3
Hyson	140 – 142	141 ± 1	0.9
Lipton	142 – 147	144 ± 2	1.6
Lipton flavored black tea	186 – 194	191 ± 4	2.1

*Values are mean ± SD (mg/kg) for n = 3, the mass of tea leaves in tea bags was calculated as mean of five tea bags.

Table 5. Fluoride level of the tea infusions for 10 min brewing time for 2 x 3 or 3 x 3 samples of each tea brand taken from one or different shops (each sample was measured three times).

Tea brands name	Range, mg F^-/kg	*Mean F^-, mg/kg	% RSD
Green tea bags			
Ethiopian green tea	238 – 245	243 ± 4	1.6
Hyson green tea	130 – 131	130 ± 1	0.3
Quality green tea	241 – 248	245 ± 3	1.4
Black tea leaves			
Abay	726 – 731	728 ± 3	0.4
Abyssinia	613 – 619	616 ± 3	0.5
Addis	332 – 334	333 ± 1	0.4
Ahadu	658 – 663	660 ± 3	0.4
Almeta	301 – 303	302 ± 1	0.2
Anbessa	525 – 530	527 ± 3	0.5
Eirmon	478 – 483	481 ± 3	0.6
Gumaro	469 – 475	472 ± 3	0.6
Haron	452 – 455	454 ± 2	0.4
Hyson	151 – 155	153 ± 2	1.6
Lipton	153 – 159	157 ± 3	2.1
Mohmood	165 – 168	193 ± 1	0.3
Wushwush	622 – 625	623 ± 2	0.3
Quality	257 – 263	261 ± 3	1.3
Black tea bags			
Addis	296 – 300	298 ± 2	0.7
Hyson	193 – 194	167 ± 2	1.1
Lipton	200 – 205	203 ± 3	1.3
Lipton flavored black tea	256 – 262	258 ± 4	1.4

*Values are mean ± SD (mg/kg) for n = 3, the mass of tea leaves in tea bags was calculated as mean of five tea bags.

After 5 and 10 min brewing time the fluoride content of the infusions follow similar hierarchy except for some interchanges with that brewed within 3 min (Table 3-5). This interchange may arise due to the differences that exist in the form of the fluoride found in the teas and higher level of infused tea fluoride was found in the Ethiopian than the imported ones again.

For the same tea brands the fluoride content of the tea bags was higher than the tea leaves in the two imported black teas (Hyson and Lipton). This could be explained by the fact that black tea bags are produced from very fine and older tea leaves which accumulate higher concentration of fluoride through time. While the black tea leaves are prepared from younger shoots of the plant which contain lesser fluoride than the leaf used to make the bag form. But such comparison was not found in the Ethiopian black tea brand Addis, which has both, leaf and bag forms. The results for this brand showed that the leaf form contain higher fluoride concentration 292, 308, and 333 mg/kg for 3, 5, and 10 min brewing time, respectively, than the bag form which has 189, 246, and 302 mg/kg for 3, 5, and 10 min brewing time, respectively. Although no effect was observed by the paper bag used to store the leaves in tea bags on the fluoride concentration of the tea infusions for other brands studied [11, 19], the paper bag used for this product may have its own effect in decreasing the amount of fluoride infused to the tea liquor which requires further investigation and validation from traceable sampling data using the same product batches.

For the Hyson tea brand, where its green tea bag (GTB), the fluoride content follow BTB > BTL > GTB (BTB: black tea bag, BTL: Black tea leaves) in all the brewing time used. This is

possibly due to the preparation of green teas from younger leaves as compared to the black tea [9, 11, 15]. In this study two types of quality tea were taken, GTB and BTL. The fluoride concentration of the green tea was lower than the black tea with mean fluoride concentration of 158, 190, and 245 mg/kg for the GTB and 180, 194, and 261 mg/kg for the BTL for 3, 5, and 10 min brewing time, respectively. For Lipton black tea, its bag form was found to contain higher fluoride than its leaf form in all the brewing times used. Among the Ethio Agri-CEFT PLC products; Ethiopian green tea, Addis tea bag, and Addis tea leaf (Gold label) which are all produced from tea leaves harvested from Wushwush and Gumaro tea farms – Ethiopia; the fluoride level found was in the order of BTL > BTB > GTB.

Fluoride extraction into tea infusions

The extraction of fluoride in to tea brew from tea leaves was compared for different types of teas and tea brands and for different brewing time (Table 6). The amount of fluoride in the examined extracts increases after 5 and 10 min of brewing. As summarized in Table 6, the release of fluoride depends on both the brewing time and the type of tea brand used.

Table 6. Average increase of fluoride content in tea liquors after 5 and 10 min brewing relative to the 3 min infusion.

Tea brands name	Mean % increment, 5 min	Mean % increment, 10 min
Green tea bags		
Ethiopian green tea	10.1	66.7
Hyson green tea	40.2	63.7
Quality green tea	20.7	55.2
Black tea leaves		
Abay	7.6	14.9
Abyssinia	2.1	11.8
Addis	5.7	14.3
Ahadu	5.3	20.5
Almeta	28.8	57.1
Anbessa	11.5	32.4
Eirmon	19.0	34.7
Gumaro	17.7	37.8
Haron	3.2	29.7
Hyson	42.4	86.0
Lipton	29.8	72.7
Mohmood	18.4	41.3
Wushwush	9.8	19.7
Quality	7.9	44.8
Black tea bags		
Addis	30.4	57.6
Hyson	13.4	55.2
Lipton	30.9	84.9
Lipton flavored black tea	33.7	80.7

Fluoride intake by human and safety evaluation of teas

Fluorine is an essential element in human diet based upon its important role in bone and teeth mineralization, stimulatory and inhibitory effects on many soft tissue enzymes and dental caries resistance. The increased fluoride intake with water and food as well as on occupational

exposure on fluoride dust could be a reason for skeletal and dental fluorosis in humans [9, 10, 12, 36, 37].

Fluoride intake threshold is a problem that should get more public attention. Several national and international organizations have already recommended standards [3, 4, 9, 10], as summarized in Table 7.

Table 7. Recommended daily fluoride intake (mg/day).

People	WHO	USEPA	China	Poland
Children	2 (WHO, 2002)	2.5 (USEPA, 1985)	2.4 (China Ministry of Health, 1997)	1.5-2.5
Adult	2-4 (WHO, 1984)	4 (USEPA, 1984)	3.5 (China Ministry of Health, 1997)	1.5-4.0

The present study revealed that fluoride level of BTL after 5 min brewing, which is the common time used to prepare tea infusions, was in the range of 117–682 mg/kg, for GTB it was between 111–190 mg/kg and that of BTB fluoride concentration was in the range of 141–246 mg/kg. While the Ethiopian black teas leaves fluoride level ranges from 248 to 682 mg/kg with mean concentration of 458 mg/kg among ten brands.

Table 8. Expected daily fluoride intake through consumption of different amount of teas from the studied brands.

Tea brands name	DFI, mg 1 cup	DFI, mg 2 cup	DFI, mg 3 cup	DFI, mg 4 cup	DFI, mg 5 cup
Green tea bags					
Ethiopian green tea	0.30	0.69	0.89	1.19	1.49
Hyson green tea	0.22	0.43	0.65	0.87	1.08
Quality green tea	0.38	0.76	1.13	1.51	1.89
Black tea leaves					
Abay	1.70	3.41	5.11	6.82	8.52
Abyssinia	1.40	2.81	4.21	5.62	7.02
Addis	0.77	1.54	2.31	3.08	3.85
Ahadu	1.44	2.89	4.32	5.77	7.21
Almeta	0.62	1.24	1.86	2.48	3.10
Anbessa	1.11	2.22	3.33	4.44	5.55
Eirmon	1.06	2.12	3.18	4.24	5.30
Gumaro	1.00	2.01	3.02	4.03	5.03
Haron	0.90	1.80	2.70	3.60	4.50
Hyson	0.28	0.56	0.83	1.11	1.39
Lipton	0.30	0.59	0.89	1.18	1.48
Mohmood	0.35	0.70	1.04	1.39	1.73
Wushwush	1.42	2.86	4.28	5.71	7.13
Quality	0.48	0.95	1.42	1.90	2.37
Black tea bags					
Addis	0.51	1.01	1.51	2.02	2.52
Hyson	0.23	0.46	0.69	0.91	1.14
Lipton	0.25	0.50	0.75	1.00	1.25
Lipton flavored black tea	0.31	0.62	0.94	1.25	1.56

DFI = Daily fluoride intake, mg.

Assuming that on average one consumes 4 cups of tea everyday (400 mL) and each cup uses 2.5 g of tea leaves or one tea bag, daily fluoride intake through BTLs could be as high as 6.82 mg and 1.38 mg in case of GTB, 1.81 in case of BTB. From the Ethiopian black tea leaves alone, the daily fluoride intake may range from 2.48–6.82 mg. Thus, according to the WHO recommendation for daily fluoride intake thresholds for different age groups, consumption of 4 cups of tea prepared from the green and black tea bags is safe for all age groups (children, adolescents and adults) considering other sources of daily fluoride intake are very minimum with respect to the fluoride found in the teas.

But in this study it was observed that consuming 4 cups of teas prepared from the Ethiopian black tea leaves will exceed the recommended daily fluoride intake of 4 mg by WHO even for adults. But some high quality teas produced in Ethiopia like Addis black tea leaf (Gold label) provide around 3.0 mg fluoride if 4 cup of tea is consumed daily, which is safer for adults but higher for children and adolescents.

Thus according to the World Health Organization [22] recommendation for daily fluoride intake threshold which is 2 mg to children and adolescents, and 4 mg for adults; and assuming 4 cups of tea daily consumption, no or very less amount fluoride from other sources; all the black and green tea bags, imported black tea leaves are safe for all age groups. From the 10 Ethiopian black tea leaf brands none of them are safe for children but 30% of them are safe for adults. But if 2 cups of tea daily consumption is considered this increases to 100% and the former to 40% and further increases to 100% if only one cup of tea daily consumption is considered. For 5 cups of tea daily consumption only 20% of the teas are safe for adults. Table 8 shows daily fluoride intake in mg through consuming different cups of teas of different brand.

Table 9. Expected fluoride intake through consumption of 4 cups (400 mL) of the teas brewed with drinking water from various areas of the country with different fluoride level.

Tea brands name	AWDWFL <0.1 mg/L	AWDWFL 0.1 – 0.4 mg/L	AWDWFL 0.4 – 1.3 mg/L	AWDWFL 1.3 – 3.5 mg/L	AWDWFL 3.5 – 7.1 mg/L
Green tea bags					
Ethiopian green tea	<1.23	1.23 – 1.35	1.35 – 1.71	1.71 – 2.59	2.59 – 4.03
Hyson green tea	<0.90	0.90 – 1.02	1.02 – 1.39	1.39 – 2.27	2.27 – 3.70
Quality green tea	<1.55	1.55 – 1.67	1.67 – 2.03	2.03 – 2.91	2.91 – 4.35
Black tea leaves					
Abay	<6.86	6.86 – 6.98	6.98 – 7.34	7.34 – 8.22	8.22 – 9.66
Abyssinia	<5.66	5.66 – 5.78	5.78 – 6.14	6.14 – 7.02	7.02 – 8.46
Addis	<3.12	3.12 – 3.24	3.24 – 3.60	3.60 – 4.48	4.48 – 5.92
Ahadu	<5.81	5.81 – 5.93	5.93 – 6.29	6.29 – 7.17	7.17 – 8.61
Almeta	<2.52	2.52 – 2.64	2.64 – 3.00	3.00 – 3.88	3.88 – 5.32
Anbessa	<4.48	4.48 – 4.60	4.60 – 4.96	4.96 – 5.84	5.84 – 7.28
Eirmon	<4.28	4.28 – 4.40	4.40 – 4.76	4.76 – 5.64	5.64 – 7.08
Gumaro	<4.07	4.07 – 4.19	4.19 – 4.55	4.55 – 5.43	5.43 – 6.87
Haron	<3.64	3.64 – 3.76	3.76 – 4.12	4.12 – 5.00	5.00 – 6.44
Hyson	<1.15	1.15 – 1.27	1.27 – 1.63	1.63 – 2.51	2.51 – 3.95
Lipton	<1.22	1.22 – 1.34	1.34 – 1.70	1.70 – 2.58	2.58 – 4.02
Mohmood	<1.43	1.43 – 1.55	1.55 – 1.91	1.91 – 2.79	2.79 – 4.23
Wushwush	<5.75	5.75 – 5.87	5.87 – 6.23	6.23 – 7.11	7.11 – 8.55
Quality	<1.94	1.94 – 2.06	2.06 – 2.42	2.42 – 3.30	3.30 – 4.74
Black tea bags					
Addis	<2.06	2.06 – 2.18	2.18 – 2.54	2.54 – 3.42	3.42 – 4.86
Hyson	<0.95	0.95 – 1.07	1.07 – 1.43	1.43 – 2.31	2.31 – 3.75
Lipton	<1.04	1.04 – 1.16	1.16 – 1.52	1.52 – 2.40	2.40 – 3.84
Lipton flavored black tea	<1.29	1.29 – 1.41	1.41 – 1.77	1.77 – 2.65	2.65 – 4.09

AWDWFL = Areas with drinking water fluoride level.

As distilled-deionized water was used for the preparation of tea infusions in this study, the actual amount of fluoride taken from tea brewed with drinking water may be elevated. The fluoride level of drinking water in many areas of the country including the capital city – Addis Ababa is less than 1.30 mg/L [38] where as in areas of the Ethiopian Rift Valley it ranges from 1.2–36 mg/L with mean concentration of 10 mg/L [36]. Though, no water sample from any part of the country was analyzed for its fluoride level in this study, Table 8 can be modified for teas brewed with drinking water of various fluoride levels from different part of the country (Table 9).

CONCLUSIONS

Fluoride levels of tea infusions prepared from 21 teas were determined by fluoride ISE method at three different brewing times. The main findings of this study are: (i) increasing brewing time increases fluoride level in the tea liquors, (ii) the Ethiopian teas were found to contain higher level of fluoride than those of imported ones and some other brands consumed abroad, (iii) for imported teas of the same brand the fluoride level was in the order of green tea < black tea leaf < black tea bag, this is due to: green teas are made from very young shoots of the tea plant than those used for black tea and in the general world market tea bags are prepared from very fine older dust tea leaves which accumulate higher fluoride level through aging, and (iv) from the safety evaluation results: assuming no other source of dietary fluoride, consumption of 4 cups (5 min brewing time) of the black and green tea bags; and imported black tea leaves are safe for all age group according to the WHO guideline for daily fluoride intake thresholds. While daily consumption of 4 cups of any of the black Ethiopian teas analyzed are unsafe for children and only 30% are safe for adults.

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