FLUORIDE CONTENT OF TEAS AVAILABLE ON THE POLISH MARKET

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ABSTRACT: The tea plant *Camellia sinensis* L. accumulates high levels of fluoride, and up to 98% of the fluoride is transferred to the leaves. This study determined the levels of fluoride in selected tea infusions in 37 different kinds of tea available on the Polish market, including 13 samples of black tea, 20 of green tea, and 4 of fruit and herbal teas. The fluoride concentration was measured potentiometrically. The results showed that the fluoride concentrations differed significantly between black and green tea infusions. The highest fluoride concentrations were found in black tea infusions, ranging from 0.728 to 3.496 mg/L. Green tea infusions contained lower concentrations of fluoride, ranging from 0.153 to 2.805 mg/L. The lowest fluoride concentrations were measured in fruit and herbal teas, ranging from 0.049 to 0.059 mg/L. In all infusions, fluoride levels increased with increasing brewing times.

Keywords**:** Brewing time; Contamination of food; Fluoride; Potentiometric method; Tea.

INTRODUCTION

Fluorine is a common element in the environment and has a impact on a large range of organisms. Although it is not an essential element, the use of the fluoride ion topically is considered to have a beneficial anticaries effect by protecting enamel and dentine as a result of its ability to stabilize the hydroxyapatite structure of enamel.[1,2] However, excess amounts of fluoride cause dental and skeletal fluorosis as well as metabolic, morphological, and functional disorders of the soft tissues of the brain, liver, kidneys, and the spinal cord.[3-6] In addition, fluoride in high doses can accelerate ageing processes and stimulate the production of reactive oxygen species, which are responsible for lipid peroxidation and oxidative stress in organisms.[7-8] Studies have shown that IQ values of children from areas polluted with fluoride are lower than those of children from non-polluted areas.[9]

The rapid development of a variety of industries has resulted in increased levels of fluoride in the environment, which, in some cases, are significantly higher than the natural background levels. Fluorine compounds are commonly used in, or formed as a by-product from, aluminum, copper, and iron smelters, and the ceramic and phosphate fertilizer industries.[10] It can therefore be safely said that the main source of fluoride in the environment, and hence in the human body, is industrial production. Apart from pollution, there are a number of other fluoride sources, such as the fluoride-enriched materials used for caries prevention, e.g., tooth pastes, tablets, rinses, and dental sealants.[11] Fluoride can also reach the human body via the uptake of plant- and animal-based foods. Although the fluoride concentrations of most fruits, vegetables, and animal-derived foods are insignificant, the levels of fluoride ions can significantly differ between different samples of the same type of food.[12] One of the plant species which absorb high amounts of fluoride is the tea plant *Camellia*

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sinensis, which can take up fluoride from the soil as well as from the atmosphere. A previous study found that up to 98% of the accumulated fluoride is stored in the leaves,[13] although the amount of fluoride in the leaves is highly dependant on the developmental stage and age of the plant. Generally, fluoride levels are higher in older plants.[14] Fluoride concentration in tea leaves usually ranges between 3.2 and 260 mg F^-/kg.[15] However, a number of studies have found considerably higher fluoride amounts in Chinese tea produced from fallen and old leaves, ranging from 708 up to 1,175 mg F^-/kg.[14,16] This should be a matter of concern, as during the process of brewing, 68.8–86.8% of fluoride gets released from the leaves into the infusion and is then absorbed by the human organism.[17] Consequently, the consumption of three cups of tea per day (each with an average fluoride concentration of 150 mg/kg) may result in an increased fluoride intake (up to 40% of daily intake) and reach a level of 4.0 mg F^- per day. However, the intake varies depending on age, sex, and body weight.[18,19]

Poland is a major importer of tea. In terms of tea consumption, the country ranks 4th in the European Union and 9th in the world.[20] In the last few years, the importation of tea has increased considerably and reached 48.8 tons in 2012.[5] Annual consumption of tea in Poland is approximately 1 kg per person.[20] Tea is usually consumed two to three times a day and sometimes even more frequently. Consequently, the daily intake of fluoride is relatively high.

The aim of this study was therefore to determine the amount of fluoride in infusions of selected teas available in Poland, derived from different plantations in different geographic locations, depending on the brewing time.

MATERIAL AND METHODS

Thirty seven samples of different teas available on the Polish market were analyzed.

Infusions were prepared by placing 2.00 ± 0.001 g of each sample into a plastic cup and pouring 100 cm^3 of deionized boiling water (100°C) over the tea sample. Brewing times were 1 and 30 minutes. After brewing, the infusions were filtered into plastic tubes and left to stand for a few minutes to cool to room temperature.

Fluoride concentration was determined potentiometrically using an ion-selective electrode (Thermo Scientific Orion 920A, USA) and TISAB III buffer. Each sample was analyzed in triplicate and the mean values were used for further analyses.

The results were analyzed statistically using Stat Soft Statistica 9.0 PL. To assess the differences between results, Tuckey's test was used. The level of significance was set at $p<0.05$.

RESULTS AND DISCUSSION

Tables 1–4 show the results obtained. Fluoride concentrations in samples of black tea differed statistically from those in other teas. The fluoride levels in black tea infusions ranged from 0.728 to 3.496 mg/L (Table 1). In this group of teas, the highest fluoride concentrations, depending on the brewing time (1 or 30 min.), were found in Saga tea (2.868 and 3.496 mg/L, respectively) and the lowest in the tea

brands Dilmah Afternoon, Tetley, Twinings, and Lipton. Brewing time had a significant influence on the fluoride contents in all samples. After a brewing period of 30 min, the highest increase in fluoride level was noticed for Tetley (192.2%).

The results of the present study were similar to those obtained by Emekli-Alturfan et al.,[21] who found fluoride concentrations ranging from 0.57 to 3.72 mg/L.

Table 1. Fluoride contents (mg F/L) in black tea infusions depending on the brewing time. (Values represent mean±standard deviation [SD])

Commercial brand name	Type of tea	Country of origin	Brewing time (mean±SD)		Increase of F content (%)
			1 min	30 min	
Lipton Yellow Label Tea	Tea bags	Sri Lanka	1.671±0.085^{c,d,B}	2.245±0.115^{e,A}	34.38
Lipton Yellow Label Tea	Granulated	Sri Lanka	1.375±0.121^{e,B}	2.759±0.485^{b,A}	99.13
Lipton Yellow Label Tea	Leaves	Sri Lanka	1.255±0.145^{e,B}	1.858±0.062^{g,A}	48.05
Twinings	Leaves	China	0.955±0.133^{f,B}	1.558±0.187^{h,A}	63.08
Tetley	Leaves	Sri Lanka	0.764±0.026^{g,B}	2.234±0.028^{e,A}	192.20
Tesco Earl Grey Strong	Tea bags	India, Kenya, Vietnam	1.541±0.041^{d,B}	2.643±0.266^{c,A}	71.53
Saga	Tea bags	China	2.868±0.354^{a,B}	3.496±0.493^{a,A}	21.91
Saga	Granulated	China	2.041±0.068^{b,B}	2.898±0.056^{b,A}	42.00
Ahmad Earl Grey	Tea bags	Sri Lanka	1.305±0.065^{e,B}	1.869±0.089^{g,A}	46.61
Teekanne - Irish Cream	Tea bags	China	1.394±0.025^{e,B}	2.084±0.162^{f,A}	49.47
Emix	Granulated	India	1.885±0.004^{c,B}	3.352±0.219^{a,A}	77.78
Royal tea z dodatkami	Tea bags	Sri Lanka	1.543±0.012^{d,B}	2.496±0.075^{d,A}	61.71
Dilmah Afternoon	Leaves	Sri Lanka	0.728±0.051^{g,B}	1.026±0.134^{i,A}	40.92

Values denoted by the same lower case (small) letter in the same columns and by the same upper case (capital) letter in the same rows or lines do not differ significantly at $p<0.05$.

Green tea, compared to black tea, contained significantly less fluoride (Tables 2 and 3), with an average fluoride concentration ranging from 0.153 to 2.805 mg/L. In the green tea group, the highest fluoride levels were determined in the brands Tetley (with orange and guarana), Vitax, and Yunnan (jasmine), while the lowest levels were found in Dakir Green and White, China Green-Chun Mee, Japan Sencha, and China Green with orange and ginger. A prolonged brewing period of 30 min resulted in increased levels of fluoride in each sample. The highest increase, by 189%, was found for Dakir Green and White tea. These results were in agreement with the findings of Zerabruk et al., who observed that the levels of fluoride in tea samples depended on the brewing time.[13]

Table 2. Fluoride contents (mg F/L) in green tea infusions depending on the brewing period. (Values represent mean±standard deviation [SD])

Commercial brand name	Type of tea	Country of origin	Brewing time (Mean±SD)		Increase of F content (%)
			1 min	30 min	
Tetley	Leaves	India	1.272±0.056^{c,d,B}	1.775±0.078^{f,A}	39.51
Tetley with orange and guaran	Tea bags	India	1.741±0.085^{a,B}	2.721±0.108^{a,A}	55.17
Herbapol with lemon	Leaves	China	0.998±0.168^{f,B}	1.814±0.031^{d,A}	81.83
Herbapol	Tea bags	China	1.330±0.092^{c,B}	1.736±0.091^{f,A}	30.49
Bio-Active with *Opuntia ficus-indica* and acacia	Leaves	China	0.847±0.105^{g,B}	1.821±0.094^{d,A}	114.90
Bio-Active with raspberries, cornflower and hibiscus	Leaves	China	1.315±0.010^{c,B}	2.396±0.009^{c,A}	82.15
Bio-Active with quince	Leaves	China	1.275±0.006^{c,d,B}	1.912±0.074^{d,A}	49.95
Bio-Active raspberries	Tea bags	China	1.382±0.036^{c,B}	1.891±0.055^{d,A}	36.73
Yunnan	Leaves	China	0.771±0.009^{g,B}	1.517±0.058^{g,A}	96.76
Yunnan jasmine	Leaves	China	1.102±0.014^{e,B}	2.560±0.073^{b,A}	132.31
Yunnan	Tea bags	China	1.521±0.072^{b,B}	1.770±0.068^{f,A}	16.37
China Green - Chun Mee	Leaves	China	0.371±0.021^{h,B}	0.701±0.013^{i,A}	88.59
Japan-sencha	Leaves	Japan	0.352±0.024^{h,B}	0.908±0.035^{h,A}	157.81

Values denoted by the same lower case (small) letter in the same columns and by the same upper case (capital) letter in the same rows or lines do not differ significantly at $p<0.05$.

Table 3. Fluoride contents (mg F/L) in green tea infusions depending on the brewing period. (Values represent mean±standard deviation [SD], EU=European Union)

Commercial brand name	Type of tea	Country of origin	Brewing time (Mean±SD)		Increase of F content (%)
			1 min	30 min	
China Green with orange and ginger	Leaves	China	0.376±0.003^{h,B}	0.597±0.051^{j,A}	58.41
Feel green (pineapple flavour)	Tea bags	India	1.303±0.075^{c,B}	2.137±0.033^{d,A}	64.05
Dakir Green & White	Leaves	India	0.153±0.017^{i,B}	0.445±0.113^{k,A}	189.59
Minutka with *Opuntia ficus-indica* and quince	Tea bags	China	1.203±0.012^{d,B}	1.664±0.087^{f,A}	38.27
Bastek with *Opuntia ficus-indica*	Leaves	China, EU	1.106±0.011^{e,B}	1.727±0.068^{f,A}	56.11
Vitax	Leaves	EU	1.576±0.044^{b,B}	2.805±0.085^{a,A}	77.94
Saga	Tea bags	Kenya, India	1.304±0.007^{c,B}	1.715±0.072^{f,A}	31.47

Values denoted by the same lower case (small) letter in the same columns and by the same upper case (capital) letter in the same rows or lines do not differ significantly at $p<0.05$.

In the infusions of the analyzed fruit and herbal teas (Table 4), the fluoride levels were the lowest, with no significant differences between the different teas. In addition, the brewing time had no impact on the fluoride levels in the infusions.

These results were similar to the findings of Giljanović et al., who observed fluoride levels below 1 mg/L in herbal tea infusions.[22]

Table 4. Fluoride contents (mg F/L) in fruit and herbal tea infusions depending on the brewing time (Values represent mean±standard deviation [SD])

Commercial brand name	Type of tea	Country of origin	Brewing time (Mean±SD)		Increase of F content (%)
			1 min	30 min	
Minutka with blackberries, raspberries, and bilberries	Tea bags	Poland	$0.063 \pm 0.002^{b,A}$	$0.068 \pm 0.023^{a,A}$	12.92
Natur-Vit with hibiscus	Leaves	China	$0.049 \pm 0.005^{b,A}$	$0.059 \pm 0,008^{c,A}$	18.79
Biofix Mentha	Tea bags	Poland	$0.061 \pm 0.006^{b,A}$	$0.061 \pm 0.009^{c,A}$	Not found
Lord Nelson – with vanilla	Tea bags	Kenya	$0.052 \pm 0.003^{b,B}$	$0.112 \pm 0.017^{b,A}$	86.66

Values denoted by the same lower case (small) letter in the same columns and by the same upper case (capital) letter in the same rows or lines do not differ significantly at $p<0.05$.

According to Czerwińska,[23] the brewing time for most types of teas should not exceed 5 minutes. Dołęgowska and Masternak have observed higher fluoride levels of tea infusions (up to 20 mg/L) with increased brewing time.[24]

Fluoride concentrations in tea infusions are also affected by the age of the leaves. Generally, older leaves have higher amounts of fluoride.[25] This explains the relatively high fluoride levels of black teas from teabags, which are usually produced using low-quality crushed leaves, stems, and tea wastes.[25] According to Zerabruk et al. and Stańczyk, the fluoride levels of old tea leaves can range between 5.30 and 23.50 mg/L.[13,18] These values were similar to those found in the present study. Generally, fluoride levels were higher in teas from teabags, irrespective of the group of the tea. In contrast, infusions from loose leaf tea did not exceed fluoride levels of 2.9 mg/L.

Another factor which influences the fluoride levels of teas is the geographical region, which could also be verified in our study. According to Buzalaf et al., teas from Turkey are characterized by higher fluoride concentrations than teas from Sri Lanka.[26] Zerabruk et al. have demonstrated relatively high levels of fluoride in Ethiopian teas.[13]

Such differences in the fluoride levels are mainly caused by the different fluoride concentrations of the surface water, varying levels of environmental pollution,[27] and the fluctuating use of fluoride-containing fertilizers in different countries.

Given the high consumption of tea in Poland, especially in the form of black tea in teabags, monitoring the levels of fluoride in teas available on the Polish market has become a necessity to ensure consumer safety.

CONCLUSIONS

In the present study the highest fluoride concentrations were found in black tea infusions (teabags and granulated tea) and lower concentrations were found in fruit tea infusions. Across the different teas, the lowest fluoride levels were measured in the loose leaf tea infusions. Fluoride concentrations of the infusions increased with brewing time. The results obtained suggest that the frequent consumption of black tea, especially when brewed for a long period, increases fluoride uptake from the diet.

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