

Assessment of fluoride concentration and daily intake by human from tea and herbal infusions

E. Malinowska ^a, I. Inkielewicz ^b, W. Czarnowski ^b, P. Szefer ^{a,*}

^a Department of Food Sciences, Medical University of Gdańsk, Hallera 107, PL 80-416 Gdańsk, Poland

^b Department of Toxicology, Medical University of Gdańsk, Hallera 107, PL 80-416 Gdańsk, Poland

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Abstract

The fluoride content in infusions of commercially available black, green, oolong, pu-erh and white teas was determined by ion-selective electrode. Herbal infusions as well as instant tea and ready-to-drink tea beverages were also examined. It is found that brewing time (5, 10 and 30 min) does increase the fluoride content, which in infusions of black tea (5 min brewing) was higher than that in the other types of tea, with contents ranging between 0.32 and 4.54 mg/l for black tea to 0.37–0.54 mg/l for white tea and with even lower values for herbal tea infusions of 0.02–0.09 mg/l. On the basis of the results obtained, the daily intake of fluoride provided from tea and herbal beverages was estimated for an adult person and for children in comparison with the Polish SAI (Safe and Adequate Daily Intake) of fluoride which is strictly attributable to ADI (Acceptable Daily Intake). The fluoride intake resulted from the regular consumption of black tea infusions was raised as compared to the other types of teas as well as herbal teas. For adult and children tea drinkers consuming five cups of black tea per day the intake of fluoride will be in the range of 8.0–303% and 12–303% of the SAI, respectively. People are often exposed to multiple sources of fluoride, such as in food, water, air and excessive use of toothpaste. The control of tea quality is important to protect human against too high uptake of this element from black tea, which is the most popular beverage. Excessive intake of fluoride with black tea, especially in the regions with its high level in the drinking water, increases the risk of dental fluorosis in children during the years of tooth development. The long-term exposure to large amounts of fluoride can lead to potentially skeletal fluorosis [WHO (1984) Fluorine and Fluorides. Environmental Health Criteria 36. WHO, Geneva].

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1. Introduction

Tea infusion is after water, the most famous beverage in the world. According to the fermentation process teas produced from the leaves of an evergreen tree *Camellia sinensis*, are classified into three main categories, i.e., green (unfermented), oolong (semi-fermented) and black (fully fermented). White tea is produced in a different way as compared to all other teas. Only special leaves, new grown buds and young leaves are selected. It retains the high concentrations of catechins which are present in fresh tea

leaves. Pu-erh tea is a fermented tea, traditionally made with tender leaves from tall and old trees. The leaves are larger than other tea leaves and have a different chemical composition. They are left green or moderately fermented before being dried. Sometimes the tea is then formed into cakes or bricks, wrapped in paper or pomelo rinds, and stored underground for several years before taking on the darker and mellower characteristics that make pu-erh tea (Yamamoto et al., 1997). Also various kinds of tea with herbal supplements are consumed very often. Infusions derived from tea with *Ginkgo biloba* or *Panax ginseng* are used additionally in the treatment and prevention of many diseases because of their purported health claims particularly or antioxidant and anticarcinogenic purposes and,

* Corresponding author. Fax: +48 58 349 3110.

E-mail address: pszef@amg.gda.pl (P. Szefer).

Table 1
Tea samples analysed and their characteristics

Lp	Tea type and name	Origin/brand	Shape/additives
<i>Black tea</i>			
1	Black Yunnan Gold	China	Leaf tea
2	Black Assam Ambagun	India	Leaf tea
3	Black Darjeeling	India	Leaf tea
4	Black Assam FOP	India	Leaf tea
5	Black High Grown	Sri Lanka	Leaf tea
6	Black Maloom	Nepal	Leaf tea
7	Black Golden Tippea	Kenya	Leaf tea
8	Black	Georgia	Leaf tea
9	Black Ceylon Gold	Sri Lanka/DILMAH	Leaf tea
10	Black	India Tetley	Leaf tea
11	Black Earl Grey	India Tetley	Leaf tea
12	Black	Posti	Bags
13	Black Assam	AHMAD TEA	Leaf tea
14	Black	Lipton	Bags
15	Black Earl Grey	Lipton	Leaf tea
16	Black Cardamom	Sri Lanka/DILMAH	Bags
17	Black Yunnan	China/ZAS-POL	Leaf tea
18	Black Vanilla	Sri Lanka/DILMAH	Bags
19	Black Ygara	Africa/Astra	Granulated
20	Black Darjeeling Himalaya	India/Astra	Leaf tea
<i>Green tea</i>			
1	Green Gunpowder	China	Leaf tea
2	Green Bancha	Japan	Leaf tea
3	Green Sencha Lemon	Japan	Leaf tea
4	Green	Vietnam	Leaf tea
5	Green	Java	Leaf tea
6	Green Haichao	China/YUNNAN Co. Ltd.	Blocks
7	Green	AHMAD TEA	Leaf tea
8	Green Gunpowder	Bio-Active	Leaf tea
9	Green	China/TEEKANNE	Leaf tea
10	Green Large leaf	China/DILMAH	Leaf tea
11	Green Moroccan mint	Sri Lanka/DILMAH	Bags/peppermint leaves 20%
12	Green Yunan	China/POSTI	Leaf tea
13	Green Ninghong	China Jiangxi	Bags/yam rhizome <i>dioscoreae</i> , senna seeds, may
14	Green Jasmine	China/DILMAH	Leaf tea/Jasmine flowers
15	Green Ginseng	China/Bio-Active	<i>Panax ginseng</i>
16	Green Ginseng	Vitax	Bags/ <i>Panax ginseng</i> , apple, mate, peppermint leaves
17	Green Ginkgo	Vitax	Bags/ <i>Ginkgo biloba</i> , apple, mate
<i>Oolong and pu-erh teas</i>			
1	Oolong	China	Leaf tea
2	Oolong	Taiwan	Leaf tea
3	Pu-erh	China	Bags
4	Pu-erh Yunnan	China/TOPTI	Leaf tea
5	Pu-erh Yunnan	China/TEEKANNE	Bags
6	Pu-erh Yunnan Pu-001B	China	Bags
<i>White tea</i>			
1	White Pai MuTau	China	Leaf tea
2	White	China	Leaf tea
3	White Pandino	China Bio-Active	Leaf tea, bamboo shoots
<i>Herbal teas</i>			
1	Yerba Mate	Brasil	Leaf <i>Ilex paraguariensis</i>
2	Yerba Mate	Argentina	Leaf <i>Ilex paraguariensis</i>
3	Rooibos	South Africa	Herbs <i>Aspalathus linearis</i>
4	Rooibos Vanilla Tea	South Africa/TEEKANNE	Bags
5	Rooibos Earl Grey	South Africa/Astra	Bags
6	Rooibos	South Africa/Bio-Active	Herbs
7	Honey bush	South Africa	Herbs <i>Cyklopia intermedia</i> sp.
8	Honey bush	South Africa/Bio-Active	Herbs

Table 1 (continued)

Lp	Tea type and name	Origin/brand	Shape/additives
<i>Instant tea</i>			
1	Instant tea drink	Krüger	Black tea extract 1.28%, honey
2	Instant tea drink	MASPEX	Black tea extract 0.14%, lemon flavour
3	Instant tea drink	MASPEX	Black tea extract 0.14%, peach flavour
<i>Ready-to-drink tea beverages</i>			
1	Ice tea lemon	Lipton	Black tea, lemon flavor
2	Ice tea green	Lipton	Green tea
3	Ice tea lemon	Nestea	Black tea, lemon flavor
4	Ice tea peach	Nestea	Black tea, peach flavor

also for cardiovascular diseases, diabetes, obesity and dental caries (Atoui et al., 2005; Moro and Basile, 2000; Wang et al., 1994; Wang et al., 2000; Rasheed and Haider, 1998; Yang, 1997; Tijburg et al., 1997; Lambert and Yang, 2003). The daily consumption of tea infusions may contribute to the dietary requirements of several essential elements such as potassium, magnesium and manganese (Ferrara et al., 2001; Fernández et al., 2002). The contents of aluminum and fluoride in tea leaves are relatively higher than those in other plants. Fluoride is selectively absorbed from the soil to the tea tree and exists in tea leaf as an anion. Especially, acidic soils are conducive to take up the increased amounts of fluoride by tea plant. It has been known that fluoride levels in leaves and stems increase with the age of the plant (Cao et al., 1998; Fung et al., 1999).

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 (Peterson, 1995). In Poland the concentration of fluoride in water is generally below 1 mg/l. However, in some regions of Poland (Malbork, Nysa, Blaszki) the fluoride level is exceeded (Czarnowski et al., 1996).

Bearing in mind that fluorine is an essential element for human with a narrow margin between the requirement and toxicity, it is justifiable to control the concentrations of this element in its rich food source, i.e., teas to evaluate exposition of some groups of people and possible impedance over human health. The aim of this paper was to perform fluoride determination, by a fluoride selective electrode, in the water extracts of different types of tea (black, green, oolong, pu-erh, white) from several regions of the world and in commercial blends, green tea with herbal additives (*Panax ginseng*, *Ginkgo biloba*, jasmine flowers, peppermint leaves), as well as in instant teas and soft ready-to-drink tea beverages. Some herbal tea infusions such as yerba mate, rooibos and honey bush were examined. The daily intake of fluoride was also evaluated for tea drinkers with reference to Safe and Adequate Daily Intake of fluoride for an adult and children.

2. Materials and methods

The fluoride content was analysed in black, green, oolong, pu-erh and white tea infusion and extracts of green tea with herbal additives.

Some herbal tea infusions such as yerba mate (*Ilex paraguensis*) from South America, rooibos (*Aspalathus linearis*) and honey bush (*Cyclopia intermedia* sp.) from South Africa were examined. Instant tea extracts and bottled ready-to-drink tea beverages containing the extracts of black or green tea were also studied. Available on the Polish market tea is imported from different countries. Table 1 presents information about type of tea, name, geographic origin or brand, shape and additives.

Tea infusions were prepared using 2 g of tea leaves, granules and blocks with 100 ml of drinking water. The tea extract was collected after 5, 10 and 30 min of brewing to check the influence of brewing time on fluoride content in water extracts.

Samples of instant tea (10 g of product) were taken to prepare a drink because of lower content of tea extract (0.14–1.28%). In the case of ready-to-drink ice tea, 200 ml of beverage was taken directly from the bottle for the analysis.

The amount of fluoride in tea infusions was determined by potentiometric method according to Frant and Ross (1966) using the fluoride ion-selective electrode (Orion) in own modification. Before analysis buffer TISAB, pH 5.2, was added to obtain the optimal pH-range for fluoride determination. Reagent blanks were analyzed together with the samples. In the case of instant and ice tea beverages ready-to-drink the saturated solution of sodium acetate was added to samples to neutralize the acid reaction from citric acid and lemon juice.

The reliability of the method was tested with the certified standard reference material DC 73351 Tea. The agreement between our results and the certified values was satisfactory, the mean recover was 92.8% and the standard deviation 7.6%.

3. Results and discussion

3.1. Fluoride concentration in tea and herbal infusions

The fluoride concentrations after 5, 10 and 30 min of brewing are listed in Table 2. The content of fluoride in black tea extracts ranged from 0.32 to 4.54 mg/l after 5 min of brewing. The highest level was obtained for the infusion of granulated black tea Ygara from Africa and the lowest one was found in extract of Black Yunnan Gold. The obtained concentrations are comparable with those reported by other authors (Siebert and Trautner, 1985; Wei et al., 1989; Gulati et al., 1993; Chan and Koh, 1996; Hayacibara et al., 2004). Higher fluoride levels were found in extracts prepared from granulated and powdered tea in bags as compared to leaf tea. This shows that leaching of fluoride to infusion from leaves is less effective. Further, there were higher fluoride levels after 10 and 30 min brewing compared with levels after 5 min.

Table 2
Fluoride concentration (mg/l) in tea and herbal infusions

Type of tea and name	Origin/brand	Time of brewing (min)		
		5	10	30
<i>Black tea</i>				
Black Yunnan Gold	China	0.32	0.53	0.81
Black Assam	India	0.79	1.01	1.12
Black Darjeeling	India	0.57	0.65	0.71
Black Assam FOP	India	1.15	1.36	1.52
Black High Grown	Sri Lanka	0.72	0.91	1.08
Black Maloom	Nepal	0.51	0.57	0.63
Black Golden Tippea	Kenya	0.93	1.39	1.49
Black	Georgia	0.89	0.99	1.07
Black Ceylon Gold	Dilmah	1.08	1.28	1.34
Black	Telety	2.26	2.49	2.69
Black Earl Grey	Tetley	2.62	3.03	3.32
Black	Posti	2.25	2.32	2.61
Black Assam	Ahmad Tea	1.43	1.59	1.75
Black	Lipton	2.76	3.28	3.42
Black Earl Grey	Lipton	1.22	1.44	1.50
Black Cardamom	Dilmah	2.13	2.29	2.45
Black Yunnan	ZAS-Pol	1.09	1.37	1.57
Black Vanilla	Dilmah	2.03	2.32	2.61
Black Ygara	Astra	4.54	6.13	6.87
Black Darjeeling Himalaya	Astra	0.82	0.91	1.15
<i>Green tea</i>				
Green Gunpowder	China	0.59	1.11	1.21
Green Bancha	Japan	1.06	1.36	1.70
Green Sencha Lemon	Japan	1.25	1.59	1.80
Green	Vietnam	0.84	1.12	1.38
Green	Java	1.83	2.14	2.52
Green Haichao	China	0.71	0.79	0.94
Green	Ahmad Tea	1.12	1.26	1.38
Green Gunpowder	Bio-Active	1.46	1.74	2.09
Green	Teekane	1.34	1.56	1.69
Green Large leaf	Dilmah	1.62	1.79	1.95
Green Yunan	Posti	0.79	0.88	0.96
<i>Green tea with herbal additives</i>				
Green Moroccan mint	Dilmah	1.25	1.42	1.70
Green Ninghong	Jiangxi	0.08	0.11	0.26
Green Jasmine	Dilmah	0.98	1.15	1.31
Green Ginseng	Bio-Active	0.68	0.72	0.82
Green Ginseng	Vitax	0.21	0.31	0.39
Green Ginkgo	Vitax	0.55	0.89	1.16
<i>Oolong and Pu-erh tea</i>				
Oolong	China	0.87	1.06	1.26
Oolong	Taiwan	1.68	2.55	2.85
Pu-Erh	China	0.43	0.57	0.58
Pu-Er Yunnan	China	0.39	0.64	0.79
Pu-Erh Yunnan	Teekane	0.92	1.04	1.07
Pu-erh Yunnan Pu-001B	China	0.53	0.61	0.74
<i>White tea</i>				
White Pai Mu Tau	China	0.37	0.47	0.56
White	China	0.42	0.49	0.59
White Pandino	Bio-Active	0.54	0.59	0.69
<i>Herbal teas</i>				
Yerba mate	Brasil	0.09	0.10	0.14
Yerba mate	Argentina	0.03	0.03	0.06
Rooibos	South Africa	0.06	0.08	0.09
Rooibos Vanilla Tea	Teekane	0.03	0.05	0.06
Rooibos Earl Grey	Astra	0.02	0.04	0.06
Rooibos	Bio-Active	0.04	0.05	0.10
Honeybush	South Africa	0.03	0.06	0.09
Honeybush	Bio-Active	0.03	0.06	0.09

The fluoride concentrations determined in green tea infusions were lower than those in black tea brews, ranging from 0.59 to 1.83 mg/l after 5 min of brewing. The highest concentration was measured in Java green tea extract. The values obtained in the present study are similar to data presented by Fung et al. (1999) and higher than those reported by Hayacibara et al. (2004). Green tea is generally produced from bud with two young leaves which contain lower levels of fluoride (Shu et al., 2003).

In green tea infusions with herbal additives the fluoride content ranged from 0.08 mg/l in Ninghong tea to 1.25 mg/l in Green Moroccan mint tea after 5 min of brewing. The contents of fluoride in oolong and pu-erh tea infusions were between 0.39 and 1.68 mg/l after 5 min of brewing. The obtained results are in an agreement with those published by Hayacibara et al. (2004) and Fung et al. (1999) for oolong and pu-erh tea infusions.

The amount of fluoride in the white tea extracts was lower than that in other types of tea with the concentration from 0.37 to 0.54 mg/l after 5 min of brewing. There are unavailable literature data concerning fluoride content in white tea.

The examined herbal tea infusions were characterized by low values of fluoride, i.e., in range of 0.02–0.09 mg/l after 5 min of brewing. The highest level of this element was determined in yerba mate brews and the lowest in rooibos Earl Grey infusion. The data obtained for yerba mate extracts are in an agreement with those reported by Hayacibara et al. (2004).

3.2. Fluoride extraction into tea and herbal infusions

The extraction of fluoride into tea brew from leaves was different from other types and brands of tea. Fig. 1 illustrates dependence of fluoride content released into infusions of black, green, oolong, pu-erh and white tea on the time of brewing (5, 10 and 30 min). Fig. 2 presents fluoride content in herbal infusions dependent on the brewing time. The amount of fluoride in the examined extracts increased after 10 and 30 min of the continuous brewing. The average increase of fluoride content released into black tea infusions after 10 min of brewing was 21% (in range from 3.1% to 66%) and 37% (from 15% to 153%) after 30 min. In the green tea infusions the increase of fluoride content after 10 min of brewing ranged from 11.0% to 88%, while after 30 min this was from 20% to 105%. The growth in fluoride level in oolong tea brews after 5 and 30 min ranged between 13% and 64%, and 16–114%, respectively.

The concentration of fluoride in tea infusions may indicate the tea quality. The tea leaves harvested in the season of early summer are superior in quality to those collected in later seasons. Fung et al. (1999) reported that brick tea containing old leaves, fallen leaves and even branches, was characterized by the highest total fluoride contents than other brands made from young leaves. According to Cao et al. (2001) high fluoride content in brick tea depends

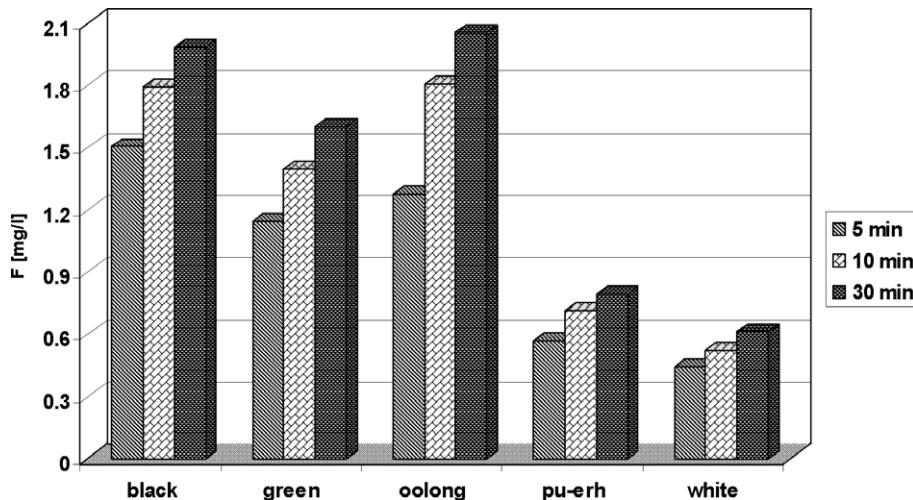


Fig. 1. Fluoride content (mg/l) in tea infusions dependent on the brewing time.

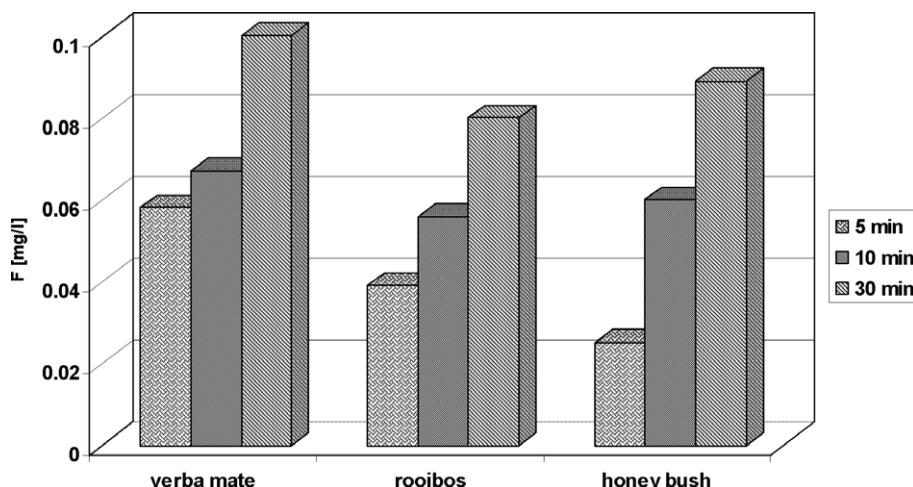


Fig. 2. Fluoride content (mg/l) in herbal infusions dependent on the brewing time.

on its content in raw material and is not caused by processing procedures.

3.3. Fluoride concentration in instant and ready-to-drink tea beverages

The fluoride levels in instant teas and ready-to-drink tea beverages are listed in Table 3. The beverages prepared from the granular instant powder of tea extract contained fluoride in range of 0.04–1.21 mg/l, while in ready-to-drink bottled ice tea beverages from 0.66 to 1.65 mg/l. According to Cao et al. (2004) the fluoride content in bottled black tea beverages was within the range of 0.70–0.96 mg/l. In case of instant tea and bottled ready-to-drink tea beverages, fluoride level depends on both the concentration in water used to prepare the infusion and in tea leaves as well as other supplements. Also the fluoride content was higher in product with increased level of tea extract. Similar results were presented by Jędra et al. (2003) and Behrendt et al. (2002).

Table 3
Fluoride concentration in instant teas and ice tea ready-to-drink beverages

Type of tea	Brand	mg F/L	mg F/200 ml
<i>Instant tea</i>			
Instant tea with honey	Kruger	1.21	0.24
Instant tea with lemon flavor	Maspex	0.04	0.01
Instant tea with peach flavor	Maspex	0.04	0.01
<i>Ready-to-drink tea beverages</i>			
Ice tea black lemon	Lipton	0.66	0.13
Ice tea green	Lipton	1.65	0.33
Ice tea black lemon	Nestea	1.33	0.27
Ice tea black peach	Nestea	1.27	0.25

3.4. Statistical evaluation of the data

Dendrogram of sampling points obtained by hierarchical clustering analysis (Euclidian distance, single linkage) is shown in Fig. 3. Two main clusters – A and B can be identified. White, oolong and pu-erh teas are grouped into

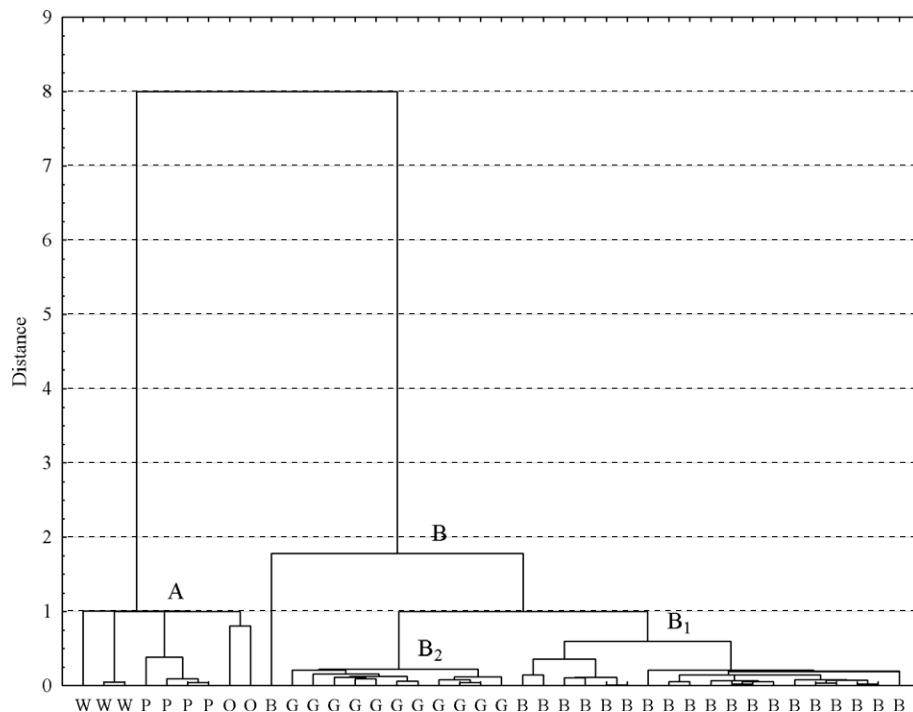


Fig. 3. Dendrogram of cluster analysis of different tea infusion samples (B – black, G – green, O – oolong, P – pu-erh, W – white).

cluster A with a similarly low levels of fluoride and originating from China. Cluster B consists of two other sub-clusters – B₁ and B₂. All the black tea infusion samples are aggregated in cluster B₂, while the green tea brews in B₁.

3.5. Fluoride intake by human

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 an occupational exposure on fluoride dust could be a reason of the skeletal and dental fluorosis in humans (Cerklewski, 1997; Wong et al., 2003).

In Poland, the SAI (Safe and Adequate Intake) is estimated for fluoride (Panczenko-Kresowska and Ziemiański, 2001). It is based closely on ADI (Acceptable Daily Intake). For an adult person the recommended safe daily intake from the all sources ranges from 1.5 to 4.0 mg and for children and teenagers, in range of 1.5–2.5 mg. In accordance with recommendations and considering an average consumption of five cups (1000 ml) per person per day, the percentage of the average daily intake of fluoride from tea and herbal extracts after 5 min of brewing was estimated. The fluoride intake from the daily consumption of black tea infusions could range from 7.9% to 303% for an adult person and from 12.8% to 303% for children. An adult tea drinker consuming five cups (1000 ml) of green tea brews per day would have an intake in range of

14.8–122% of SAI. In the case of children the green tea infusions supply 23.6–122% of SAI for fluoride.

The lowest intake of fluoride could be realized with the white tea brews which provide 9.8 for an adult person and 15.6% for children to 112% of SAI for both.

Based on fluoride concentration in the studied ice tea ready-to-drink, one bottle (500 ml) of beverage provides of 8.3% and 13.2% to 55% SAI for an adult and children. Hence, there is a risk of increased fluoride intake from these products and other sources at the same time.

In the case of herbal tea infusions, the daily intake of fluoride for adult and children with five cups (1000 ml) of their infusions can be ranged from 0.5% and 0.8% to 6.0% for an adult and for children, respectively. These results show that consuming herbal tea infusions does not influence significantly on the daily fluoride intake and these infusions may be consumed several times per day with no harmful side-effects from fluoride.

Among the analysed tea infusions, black tea extracts, especially commercial blends of different tea companies contained the highest amounts of fluoride. They are prepared by mixing of raw black teas harvested in many countries and containing different quality material such as old leaves, branches or fallen leaves.

Based on the data obtained it is concluded that consumers drinking some types of tea infusions in large quantities may be exposed to a high amount of fluoride from the drinking tea. On the other hand, herbal infusions containing the lowest fluoride concentrations can be safely consumed by adult and children, assuming that intakes from other sources remain within the SAI.

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