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FLUORIDE CONTENT OF COMMERCIALLY AVAILABLE GREEN TEA IN GERMANY AND ASSESSMENT OF THE NON-CARCINOGENIC RISK

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ABSTRACT: Green tea is consumed worldwide and the levels of fluoride ion (F), both organic and inorganic, in the product are highly important for the health of consumers. In the present study, determinations were made, for the common green tea bag brands available in Germany, of the fluoride content, the chronic daily intake, and the non-carcinogenic risk. The levels of F in a total of 15 green tea samples were analyzed by using an ion-selective electrode. The mean F content in the green tea samples was 0.704 mg g^{-1} with a range of 0.504–1.018 mg g^{-1}. The chronic daily intake (CDI) and hazard quotient (HQ) values in brand B of the green tea samples were higher in the 35–50 year-old consumers compared to the other age groups. The HQ values in the green tea consumers of the different age groups indicate that there is not a significant risk to health due to green tea consumption (HQ<1). Although tea consumers may not be exposed to adverse health effects of F from green tea, it is still important to evaluate the total F intake from different sources in order to have a better understanding of the effects of F in the human body.

Keywords: Fluoride; Green tea; Hazard quotient; Risk assessment.

INTRODUCTION

The fluoride ion (F) is a ubiquitous anion in various samples from the environment, and various industries, beverages,[1-5] and foods.[6,7] An excessive intake of F for a prolonged duration may lead to dental, skeletal, and non-skeletal fluorosis, such as thyroid function impairment.[8-12] A higher risk of dental caries was reported in children with a high consumption of sugar-containing beverages.[13] Severe adverse effects due to fluorosis have been observed in adults consuming brick tea.[14]

Tea, next to drinking water, is one of the most popular beverages consumed in the world. Tea is considered to be a refreshing beverage, which is rich in caffeine and which has proven beneficial effects for human health.[15,16] The tea plant is capable of the uptake of environmental F from the soil and the air[17,18] with up to 98% of it accumulating in the leaves,[19] which are usually used to make green or black tea. The F in tea leaves can be released easily during infusion and the daily consumption of tea can increase total F intake in humans.[20] Green tea is one of the most popular and

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common types of tea with beneficial effects on human health. [21,22] A high F content has been reported in green tea samples in China and Japan.[23] Malinowska et al. measured the F content of green tea after brewing for different time periods.[17] In another study, the F contents of popular non-, semi-, and fully-fermented green tea samples were determined in Taiwan and the results showed acute health effects in adult and children after consuming leaf or bagged green tea drinks.[24] Lu et al. investigated the F content in fresh leaves collected from different plantations of tea in China and reported a significant correlation between the F level in the different tea samples and the quality grades.[25]

Drinking tea is common source of F entry to the human body. However, only a few studies have determined the F content of green tea. Thus, the present study aimed to: (i) investigate the F contents in the different types of green tea bags that are available in the Germany market; and (ii) assess the non-carcinogenic human health risks from exposure to F via green tea consumption.

MATERIALS AND METHODS

A stock F solution of 1000 mg L^{-1} was prepared by dissolving 2.21 g NaF in 1 L of distilled water. To prepare Total Ionic Strength Adjustment Buffer (TISAB), 58 mL of glacial acetic acid and 12 g of sodium dehydrate were added to 300 mL of distilled water and stirred to obtained a homogeneous solution. Finally, the pH of the solution was adjusted to 5.2 by using NaOH and then diluted to 1 L.

Sodium hydroxide solution was prepared by dissolving 670 g NaOH pellets in distilled water. Then the solution was diluted to 1 L and stored in polyethylene containers until using for the sample preparation. All chemical materials were purchased from Merck, Darmstadt, Germany.

A total of 5 brands of green tea bag samples were selected to determine F contents. From each brand of green tea, 3 samples (total number of samples = 15) were collected from the local market in Wiesbaden, Germany, during September 2020. All samples were transferred to the laboratory and the labeled information of the green tea bag samples was recorded. Before analysis, the samples were dried at 105 ºC for 1 hr and then homogenized. The homogenized samples were passed through 40 mesh sieves and 0.5 g of each prepared sample was transferred to a 130 mL nickel crucible. Then, a little distilled water was added to the samples to moisten them. Next, the solution was prepared by dissolving with 6 mL sodium hydroxide. After that the samples were dried for one hour at 150 ºC. Dried solution was placed in a muffle furnace at 300 ºC; then, the temperature was raised to 600 ºC and samples remained at this temperature for 30 minutes. The samples were then removed from muffle furnace and allowed to cool; after that 10 mL of distilled water was added to them. The samples were heated slightly until dissolution of the sodium hydroxide fusion cake occurred and then HCL was slowly added to adjust the pH to 8–9. During this process for monitoring pH values, Fisher Alkacid Test Ribbon was used. After that the samples were transferred to a 100 mL volumetric flask and diluted to volume. The samples were passed through a dry Whatman No. 42 filter paper. Representative blanks were also prepared through the same procedure for green tea samples. The prepared samples were mixed at a 1:1 ratio with TISAB prior to analysis by an ion- selective electrode (model 781 pH/Ion meter, Metrohm, Switzerland). F measurement of each sample was repeated 3 times

and the mean values are reported here. The measurements are expressed here in mg of F per g of dry green tea (mg g^{-1}).

Human health risks were assessed from exposure to F via green tea consumption based on a Hazard Quotient (HQ). An HQ value of less than 1 implies that the daily intake of the sample, even for a long period of time, is unlikely to cause adverse health effects. Average daily exposure to F due to green tea consumptions and HQ values were evaluated based on the equations as follows:[26]

$$HQ = \frac{ADD}{RFD} \qquad \text{Equation 1}$$

$$HQ = \frac{C \times IR}{BW} \qquad \text{Equation 2}$$

Where:

ADD = the average daily dose of F via green tea (mg kg^{-1} day^{-1})

RFD = the daily oral intake reference dose of F (0.06 mg kg^{-1} day^{-1}); as suggested by the United States Environmental Protection Agency (USEPA)

C = the mean F content in green tea (mg kg^{-1}, dry weight)

IR = the consumption rate of green tea (g $person^{-1}$ day^{-1})

BW = the average body weight (kg)

The F risk assessment in green tea was evaluated for different age groups with different daily average consumption of tea infusion based on available data (Table 1).[27]

Table 1. The daily consumption of tea and average body weight of different age groups[26]

Age group (years)	Average body weight (kg)	Daily consumption (L day^{-1})
2–3	15.15	0.077
4–8	23.9	0.069
9–13	42.6	0.087
14–18	62.75	0.117
19–24	70.5	0.283
25–34	75.45	0.501
35–50	78.33	0.659
51–64	79.27	0.621
65–80	76.73	0.511

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RESULTS AND DISCUSSION

The mean (± SD) and range of the F content in the 5 different brands of green tea bag are shown in Table 2.

Table 2. Fluoride levels in green tea samples in the present study

Brand (n=3)	Range (mg g^{-1})	Mean ± SD (mg g^{-1})
A	0.654–0.812	0.731 ± 0.056
B	0.504–1.018	0.761 ± 0.229
C	0.516–0.808	0.677 ± 0.12
D	0.532–0.804	0.635 ± 0.098
E	0.608–0.776	0.712 ± 0.059
Total	0.504–1.018	0.704 ± 0/048

The F content of green tea bag samples was in the range of 0.504–1.018 mg g^{-1} with a mean value of 0.704 mg g^{-1}. The highest mean F content in the green tea samples was detected in the brand B; whereas the lowest mean F content was detected in the brand D.

In a study in China, the mean content of F in a total of 266 green tea samples was 0.063 mg g^{-1} which was lower than the results of this study.[28] In another study the F content of well-known green tea in Slovenia and in most of Europe was in the range of 0.053–0.293 mg g^{-1}.[29] In a further study on the F levels of different traditional, green, and herbal teas consumed across the world, the F content of the green tea was in the range of 0.296–1.112 mg g^{-1} and was nearly the same as in our study.[30] In another study, it was reported that an extremely high F content was released from brick tea (7.34 mg L^{-1}) compared to black tea (1.89 mg L^{-1}) and green tea (1.60 mg L^{-1}). Also, the F level in the fallen leaves (0.6–2.8 mg g^{-1}) was higher than that in fresh leaves (0.3–1.0 mg g^{-1}).[31]

Different levels of F in tea can be due to differences in the tea plant type, different soils that tea plants were grown in it, quality of water used for irrigation, type of leaf harvested, etc.[31-34]

The F levels of the green tea samples in this study are compared with other former studies as shown in Table 3.

Exposure to the excessive F from drinking tea can have detrimental effects on human health. So in this study for assessing the risk levels, the chronic daily intake (CDI) of F, in green tea consumers with different age groups, was determined. The CDI values for F via green tea consumptions in the different age groups are shown in Table 4. The CDI values for green tea ranged from 0.006–0.032 mg kg^{-1} day^{-1} (Table 4). Based on the results, the highest amount of CDI was related to the brand B of green tea consumers with 35–50 age. As tea is prepared with water containing

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different levels of F [35-39], the F intake of tea consumer will be the sum of the F in the tea as well as in the water. If we consider a range level of 0.5–1.5 mg L^{-1} for the water F level, then the estimated CDI and HQ values will be in the ranges of 0.007–0.045 mg kg^{-1} day^{-1} and 0.11–0.74, respectively.

Table 3. The fluoride contents in tea samples reported in previous studies.

Type of tea	Number of samples (n)	Mean (mg g^{-1})	Range (mg g^{-1})	References
Stick-shaped black tea	5	-	0.097-0.148	40
Granular black tea	8	-	0.139-0.223	
Beverage black tea	3		0.053-0.072	
Different teas	36	-	0.1-0.451	41
Flavored green tea	15	-	0.015-0.185	42
Black tea	15	-	0.296-0.797	30
Green tea	9	-	0.296-1.112	
White tea	1	0.546 ± 0.013	-	
Oolong tea	4	-	0.393-0.744	
Puerh tea	3	-	0.523-0.692	
Black tea	12	-	0.068-0.435	29
Green tea	7	-	0.053-0.293	
Oolong tea	2	-	0.123-0.197	
Puerh tea	3	-	0.083-0.235	
Stick black tea	6	0.125 ± 0.008	0.024-0.035	43
Granular black tea	6	0.154 ± 0.028	0.018-0.061	
Black tea bag	25	0.285 ± 0.026	0.029-0.15	
Black tea	60	0.035-0.182	0.012-0.195	44
Tea liquor	60	0.027-0.13	-	
Green tea	**15**	**0.704±0.048**	**0.504-1.018**	**Present study**

Table 4. Chronic daily intake (CDI) and Hazard Quotient (HQ) induced by fluoride exposure via green tea consumption.

Age category	CDI (mg kg^{-1} day^{-1})					HQ			
	A	B	C	D	E	A	B	C	D
2-3	0.019	0.019	0.017	0.016	0.018	0.31	0.32	0.29	0.27
4-8	0.011	0.011	0.010	0.009	0.010	0.18	0.18	0.16	0.15
9-13	0.007	0.008	0.007	0.006	0.007	0.12	0.13	0.12	0.11
14-18	0.007	0.007	0.006	0.006	0.007	0.11	0.12	0.11	0.10
19-24	0.015	0.015	0.014	0.013	0.014	0.25	0.26	0.23	0.21
25-34	0.024	0.025	0.022	0.021	0.024	0.40	0.42	0.37	0.35
35-50	0.031	0.032	0.028	0.027	0.030	0.51	0.53	0.47	0.45
51-64	0.029	0.030	0.027	0.025	0.028	0.48	0.50	0.44	0.41
65-80	0.024	0.025	0.023	0.021	0.024	0.41	0.42	0.38	0.35

The HQ values via F exposure in green tea samples were also evaluated and are presented in Table 4. The risk estimates showed that the highest values of HQ in green tea (0.53) were related to the brand B consumers of green tea in the 35–50 year- old age group. The HQ values in the green tea samples indicated that drinking tea consumers in the different age groups were not exposed to an immediate health risk (HQ<1).

In a study,[34] the exposure and risk assessment for F and different trace metals in black tea consumers with aged 8–79 year-olds were investigated.[45] The results showed that although the F content in the tea samples was greater than that of several trace metals, the HQ value of F was lower than 1 indicating that the F content in the black tea did not pose a significant human health risk.[34] In another study, the non-carcinogenic risks of F in traditional, green and herbal teas were investigated for children (<15 years) and adults (>35 years). The results showed that five green tea samples had CDI values of F higher than 0.05 mg kg^{-1} day^{-1}; although the HQ values for child and adult consumers of the different teas were lower than 1 and posed no risk of adverse health effects.[30] In another study in Iran, the CDI value of F in common black tea for men and women (21–72 years) and children (0–11 years) was evaluated. The results showed that the non-carcinogenic health risks in the age groups considered were in the order of men>women>children. In 20 percent of the cases in the men groups, a probable risk of fluorosis (HQ>1) was determined; butthe results in the other age groups showed no significant risk for tea consumers.[46]

Beside green tea, there are other various sources of F such as drinking water,[47-50] different foods,[51] various beverages,[52-53] etc. that should be considered to assess the total risk of the daily F intake.

CONCLUSIONS

In this study, the F concentration in 5 brands of green tea bag available in the Germany market place were determined and the health risks of F via green tea consumption were evaluated. According to the results, the highest values of CDI, and HQ were found in the brand B of green tea in the age group of 35–50 years old. The HQ values in all age groups were lower than 1 and this indicates that there are no immediate health effects for green tea consumers. However, it should be noted that people may also have an intake of F from other various sources, such as different foods and beverages, and that these may contribute to the total intake of F in the body and result in a high total F intake. Therefore, the intake of F from all the various sources should be considered in order to fully understand possible adverse health effects of F from the long-term consumption of green tea.

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