Environmental Pollution 231 (2017) 779–784

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Fluoride concentrations in traditional and herbal teas: Health risk assessment☆

Suchismita Das [a, b], Letuzia M. de Oliveira [a], Evandro da Silva [a], Yungen Liu [c], Lena Q. Ma [a, d, *]

[a] Soil and Water Science, University of Florida, Gainesville, USA
[b] Life Science and Bioinformatics, Assam University, Silchar, India
[c] Faculty of Environmental Science and Engineering, Southwest Forestry University, Yunnan 650224, China
[d] State Key Laboratory of Pollution Control and Resource Reuse, School of the Environment, Nanjing University, Jiangsu 210023, China

ARTICLE INFO

Article history:
Received 18 July 2017
Received in revised form
21 August 2017
Accepted 21 August 2017
Available online 30 August 2017

Keywords:
Tea
Herbal
Fluoride
Hazard quotient
Infusion

ABSTRACT

Traditional tea (*Camellia sinensis*) and herbal tea are being consumed across the world. However, long term consumption of tea can increase the chances of fluorosis owing to the presence of fluoride (F) in teas. Therefore, it is imperative to assess the health risk associated with tea consumption. The main objectives of this study were to: 1) estimate total F in 47 popular teas, including traditional and herbal teas and F concentrations in 1% (w/v) infusion of 5 min, and 2) assess the exposure risks of F from tea consumption in children and adults. The data showed that total F was the least in herbal teas (33–102 mg/kg) and their infusions (0.06–0.69 mg/L) compared to traditional teas (296–1112 mg/kg) and their infusions (1.47–6.9 mg/L). During tea infusion, 6–96% and 18–99% of the F was released into the water from herbal and traditional teas, respectively. Ten samples of traditional teas, including five green teas had chronic daily intake (CDI) values of F > 0.05 mg/d/kg bw, the stipulated permissible limits of F intake from all sources. Although the F from teas posed no immediate health hazards with hazard quotient <1, some tea samples could potentially contribute >4 mg F/d, thereby adding to the overall F burden. Therefore, together with F from food and water sources, daily F consumptions from teas might increase its health risks to humans. So, caution should be excised when drinking teas containing high F.
Published by Elsevier Ltd.

1. Introduction

Tea (*Camellia sinensis*) is a popular beverage, consumed by people worldwide (Gramza-Michałowska et al., 2016). It is rich in polysaccharides, caffeine, polyphenols and amino acids as well as antioxidants, which are beneficial for human health (Gramza-Michałowska et al., 2016; Yang and Landau, 2000). Apart from traditional tea, herbal teas are also been marketed. They often contain one or multiple herbs, so their infusions may not have *C. sinensis* leaves. Such teas are also popular amongst health-conscious people as they are rich in minerals and antioxidants (Atoui et al., 2005).

While the beneficial effects of tea are well known, pollutants in teas may pose health risk to tea drinkers (Salahinejad and Aflaki, 2010). One such pollutant is fluoride (F) in teas. In fact, tea plants are known F accumulators as soil F is readily translocated to tea leaves (Ruan et al., 2004). In tea leaves as well as tea granules, high F contents have been documented from teas in China (Lv et al., 2013), Tibet (Cao et al., 1996), Iran (Mahvi et al., 2006), Poland (Malinowska et al., 2008) and Turkey (Sofuoglu and Kavcar, 2008). In herbal infusion, high F concentrations have also been reported (Martín-Domingo et al., 2017; Kalny et al., 2007). For example, F concentrations at 0.02–0.09 mg L^{-1} in herbal infusions were reported from teas in Poland (Malinowska et al., 2008).

F in low doses is beneficial for teeth and bone health as it strengthens the apatite matrix of skeletal tissues and teeth (Barbier et al., 2010). In fact, 40–90% F in tea leaves is soluble and comprises a major source of dietary F in tea drinkers (Quock et al., 2012). However, excess F causes dental and skeletal fluorosis from chronic consumption (Ayoob and Gupta, 2006). The WHO's permissible limit for F in drinking water is 1.5 mg L^{-1} (WHO, 2011) whereas adequate F intake from all sources, including water, beverages and

☆ This paper has been recommended for acceptance by David Carpenter.

* Corresponding author. Soil and Water Science, University of Florida, Gainesville, USA.
E-mail address: lqma@ufl.edu (L.Q. Ma).

http://dx.doi.org/10.1016/j.envpol.2017.08.083
0269-7491/Published by Elsevier Ltd.

diet, is at 0.05 mg/d/kg bw (DRI, 1997). However, often the value is being exceeded due to dietary habits including drinking tea.

The exposure of tea drinkers to F is based on F content in tea as well as tea consumption rate. In oriental countries, almost a liter of tea is consumed daily by an average adult. Thus, high F in traditional and herbal teas can be of concern. The present study, thus, aimed at analyzing representative tea samples for F in total concentrations as well as in 5-min infusion in boiling water. The associated health risk to tea drinkers were estimated with reference to Safe and Adequate Daily Intake of F for adults and children.

2. Material and methods

2.1. Collection of tea samples

A total of 47 tea samples were collected from 13 countries, including 15 herbal mixes, 15 black teas, 9 green teas, 4 oolong, 3 puerh teas and 1 white tea. The F content was estimated both in total concentrations and in 5-min infusions. Table S1 presents information about tea samples, including type, country, brand, shape and ingredients (Supporting information).

2.2. Total F contents in teas and 5-min tea infusion

Tea granules were oven dried at 75 °C for 2 days. Samples for F estimation were carried out following the method of Malde et al. (2001). The method involves fusion of tea samples with 8 M NaOH at 600 °C for 30 min. The fused samples were extracted with boiling distilled water and the solution pH was adjusted to 4.25–4.75 for optimum analysis. Samples were then analyzed for F using the SPADNS colorimetric method (USEPA Method 13A, Mitchell et al., 1977; Das et al., 2017). After the F reacts with zirconium dyes, a colorless complex anion and a dye are produced. The complex, which is proportional to F concentration, tends to bleach the dye, which consequently becomes lighter with time as F concentration increases. After F reaction with SPADNS Zr^{-}, the subsequent solution color at 570 nm was determined using a spectrophotometer (Lambda 35 UV-VIS, Perkin Elmer, USA). A calibration curve using concentrations from 0 to 1.4 μg F mL^{-1} was prepared.

500 mg of oven-dried tea powder was infused with 50 mL double-distilled water at 100 °C (1%, w/v tea infusion) in a 100-mL conical flask (Fung et al., 1999). After 5-min of infusion, the tea solution was filtered through a Whatman No. 40 filter paper, pH was adjusted to 4.25–4.75 for optimum F analysis and collected in a 50-mL graduated tube. The level of F was estimated as described by Malde et al. (2001).

2.3. Quality control and assurance

All glassware were soaked overnight in 5.0 M HNO_3, rinsed with deionized water before use. The F solutions were prepared in double distilled water by diluting the prepared stock solution (1000 mg/L) to desired concentrations. Standard curve was elaborated (n = 10) using various concentrations of NaF and the measured value was within the recovery range of 97.5 ± 4.6%. In addition, samples with known F concentrations were run every 10 samples to monitor the stability of the method. The range of this method was 0–1.4 μg F/mL. All chemicals, reagents, NaF, HNO_3 and NaOH were of analytical grade (Merck, USA).

2.4. Health risk assessment and statistical analysis

In this study, exposure risk of F from tea ingestion was assessed. The daily exposure of F from ingestion was estimated following USEPA (1992):

$$CDI = C \times DI/BW \quad (1)$$

Where, CDI is the chronic daily intake (mg/kg/d), C is the F concentration in the tea infusion (mg/L), DI is the average daily intake rate of tea (L/d), and BW is body weight (kg). For children and adults, default body weight was 20 kg and 70 kg, respectively. Multiplication of C and DI is the daily F intake (mg/d).

Risks from F ingestions from teas were estimated in both children and adults. As per Sofuoglu and Kavcar (2008), an estimated of 0.075 L tea/d and 0.75 L tea/d were consumed by children < 15 years and adults > 35 years, respectively. Furthermore, the percentage of soluble F in infusions was used to interpret CDI values.

The hazard quotient (HQ) was calculated to estimate F risks using following equation (USEPA, 1999):

$$HQ = CDI/\ RfD \quad (2)$$

Where, RfD is the reference dose of F (mg/kg/d). A HQ value of >1 implies a significant risk level. For children, the risk of dental fluorosis was considered while the risk of skeletal fluorosis was considered in adults (USEPA, 1999).

All data are expressed as means of three replicates with standard error. Analyses of variance (ANOVA) by Tukey's multiple grouping were used to determine significance differences among different teas. All statistical analyses were performed with SAS statistical software (version 9.1.3, NC, USA).

3. Results and discussions

This study was embarked to estimate F contents in 47 popular traditional and herbal teas as well as to estimate the health risks of F associated with drinking teas. We carried F estimation in both total concentrations and its 5-min infusion in boiling water. The associated health risk of F to tea drinkers was estimated with reference to Safe and Adequate Daily Intake at 0.05 mg/d/kg bw (DRI, 1997).

3.1. Fluoride concentrations in total and 5-min infusion

Tea is arguably the most popular beverage, relished around the world. Teas are made from the leaves of *C. sinensis*, a species of flowering plant belonging to the family Theaceae. Based on various manufacture and processing steps, traditional tea can be divided into black (fermented), green (non-fermented), oolong (partially fermented), puerh (prolongly fermented) and white (un-oxidized) (Jain et al., 2013). Herbal tea may or may not possess *C. sinensis* leaves (Atoui et al., 2005). Typically, tea is consumed after infusing tea leaves for a few minutes using hot water (Fung et al., 1999). Therefore, we adopted 5 min of 1% (w/v) infusion of tea to estimate its F contents. F in total concentrations and in infusions are shown in Table 1. For all teas, its pH in infusion was acidic, being 3.5–5.5. Tea is cultivated in acidic soils and is known to accumulate F (Ruan et al., 2004). For all teas, the levels of F were the lowest for herbal tea at 33–102 mg/kg, followed by green tea at 297–1112 mg/kg, black tea at 296–797 mg/kg, oolong tea at 393–744 mg/kg, puerh tea at 523–692 mg/kg, and white tea at 545 mg/kg (Table 1). Similarly, F contents in infusions for herbal tea, green, black, oolong, puerh and white tea were 0.69, 6.9, 5.45, 5.6, 4.9 and 5.4 mg/L, which represented 6–96% soluble F, i.e., F was very soluble excluding herbal tea.

Tea has a long history of elevated F contents, ranging from 0.70 to 6.01 mg F/L in black teas (Cao et al., 2006). Similarly, F contents in black tea infusions from Turkey, Sri Lanka, Kenya and India are

Table 1
Fluoride in tea samples and infusion, and pH and percent of F in tea infusion.

Tea Brand	Tea type	F in tea (mg/kg)	F in 1% (w/v) infusion (mg/L)	% of F infused	pH of infusion
Yogi	Herbal	73.3 ± 3.3^{bc}	0.17 ± 0.02^{ef}	24.1 ± 1.7	5.5 ± 0.12
Stash Premium		35.0 ± 4.2^{hi}	0.25 ± 0.05^{cde}	74.8 ± 24.6	5.3 ± 0.23
Traditional medicine		33.1 ± 3.3^{hi}	0.23 ± 0.02^{def}	70.7 ± 15.3	5.1 ± 0.22
Lipton		49.1 ± 5.3^{fg}	0.46 ± 0.03^{b}	96.5 ± 17.3	5.1 ± 0.12
Herbal tea		36.9 ± 3.9^{hi}	0.33 ± 0.02^{c}	90.1 ± 3.5	5.3 ± 0.11
Boldus		44.9 ± 0.9^{gh}	0.25 ± 0.02^{cdef}	56.4 ± 3.4	5.4 ± 0.12
Runa		60.0 ± 1.7^{ef}	0.26 ± 0.04^{cde}	43.4 ± 7.5	5.5 ± 0.12
Matte Leao		61.1 ± 1.9^{de}	0.17 ± 0.01^{ef}	28.7 ± 3.0	5.5 ± 0.13
Carcueja		56.0 ± 4.7^{efg}	0.15 ± 0.03^{fg}	27.3 ± 3.5	4.8 ± 0.09
Maracuja Farms		102 ± 3.0^{a}	0.06 ± 0.02^{g}	6.0 ± 1.4	4.9 ± 0.13
Carmencita		56.0 ± 2.9^{efg}	0.31 ± 0.02^{cd}	55.3 ± 0.62	5.1 ± 0.14
Bioherbal		64.8 ± 6.4^{cde}	0.44 ± 0.05^{b}	68.3 ± 1.59	5.4 ± 0.11
Clipper		76.5 ± 3.2^{b}	0.46 ± 0.05^{b}	60.3 ± 4.14	5.3 ± 0.11
Tajo		71.9 ± 4.9^{bcd}	0.69 ± 0.03^{a}	96.7 ± 2.48	5.3 ± 0.10
Tonghai		34.3 ± 2.4^{hi}	0.22 ± 0.02^{def}	65.5 ± 12.0	5.5 ± 0.15
Tajo	Black tea	797 ± 14.3^{a}	1.47 ± 0.19^{h}	18.5 ± 2.1	4.6 ± 0.12
Stash Premium		298 ± 8.3^{j}	2.71 ± 0.26^{efg}	90.9 ± 6.3	4.8 ± 0.15
Twinings		377 ± 7.9^{i}	3.44 ± 0.48^{bcd}	91.3 ± 10.7	4.3 ± 0.13
Cha Preto Leao		668 ± 10.2^{e}	2.24 ± 0.27^{g}	33.6 ± 3.5	4.8 ± 0.12
Lipton (Yellow Label)		679 ± 1.0^{e}	2.94 ± 0.19^{def}	43.4 ± 2.8	4.3 ± 0.12
Brooke bond		703 ± 3.4^{cd}	2.69 ± 0.13^{efg}	38.3 ± 1.7	4.1 ± 0.09
Lipton (Yellow Label)		625 ± 5.0^{f}	2.38 ± 0.17^{fg}	38.0 ± 2.4	4.2 ± 0.07
Lapsang Sauchang		760 ± 0.53^{b}	3.84 ± 0.13^{bc}	50.5 ± 1.7	4.0 ± 0.12
Brooke Bond (Tajmahal)		760 ± 2.1^{b}	3.29 ± 0.19^{cde}	43.4 ± 2.3	4.1 ± 0.11
Lipton (Yellow Label)		753 ± 10.3^{b}	2.86 ± 0.27^{defg}	37.9 ± 3.0	4.3 ± 0.11
Kte (Kanchanjanha)		525 ± 4.4^{g}	4.0 ± 0.07^{b}	76.1 ± 0.8	4.1 ± 0.12
Twinings (Breakfast tea)		683 ± 2.6^{de}	5.25 ± 0.15^{a}	76.9 ± 1.9	4.3 ± 0 .012
Twinings (Darjeeling)		721 ± 7.5^{c}	5.45 ± 0.10^{a}	75.5 ± 0.59	4.4 ± 0.11
Garden Eagle		296 ± 4.9^{j}	2.66 ± 0.30^{efg}	89.9 ± 8.6	4.6 ± 0.13
African Pride		476 ± 4.6^{h}	3.49 ± 0.24^{bcd}	73.3 ± 4.3	4.7 ± 0.12
Lipton	Green tea	296 ± 9.0^{f}	2.43 ± 0.23^{e}	81.9 ± 5.4	5.3 ± 0.12
West lake Long Jing		1112 ± 12.5^{a}	3.68 ± 0.29^{d}	33.1 ± 2.2	3.8 ± 0.08
Chayuchanxin		623 ± 9.2^{d}	5.02 ± 0.15^{c}	80.6 ± 3.6	3.8 ± 0.12
Wuyutai tea		544 ± 9.9^{e}	4.59 ± 0.30^{c}	84.5 ± 7.1	4.1 ± 0.09
Huangshan meofeng		532 ± 11.0^{e}	5.31 ± 0.21^{b}	99.7 ± 1.8	4.1 ± 0.07
West lake Long Jing		600 ± 2.1^{d}	5.12 ± 0.14^{c}	85.3 ± 2.6	4.2 ± 0.12
Revolution tea		1057 ± 22.2^{b}	3.75 ± 0.22^{d}	35.5 ± 1.3	4.8 ± 0.13
Bigelow		779 ± 9.3^{c}	6.94 ± 0.08^{a}	89.1 ± 0.05	4.7 ± 0.12
Yogi		758 ± 10.0^{c}	4.94 ± 0.17^{c}	65.2 ± 1.3	4.5 ± 0.11
Lingfeng	Oolong	393 ± 10.0^{c}	3.14 ± 0.14^{c}	79.9 ± 1.5	3.9 ± 0.14
Tie Yin		421 ± 18.0^{c}	3.08 ± 0.11^{c}	73.2 ± 5.7	4.3 ± 0.15
Gaoshan (Tangji)		744 ± 10.9^{a}	5.63 ± 0.23^{a}	75.6 ± 1.9	4.4 ± 0.13
Dongdingwulong		696 ± 3.9^{b}	4.60 ± 0.15^{b}	66.2 ± 1.7	3.8 ± 0.09
Wuyutai tea	White	546 ± 13.2	5.39 ± 0.29	98.9 ± 7.7	4.6 ± 0.12
Wuyutai tea	Puerh	692 ± 9.6^{a}	4.09 ± 0.11^{b}	59.1 ± 2.4	4.4 ± 0.11
Mast Chang		523 ± 9.8^{b}	4.96 ± 0.15^{a}	94.9 ± 1.1	4.1 ± 0.11
Madui ZhanG		657 ± 19.9^{a}	2.87 ± 0.11^{c}	43.7 ± 0.35	3.9 ± 0.08

All values are mean ± SD of 3 replicates; values of different teas (total or infusion) within a particular species followed by the same letters are not significantly different at $p < 0.05$.

0.57–3.72 mg/L after 5 min brewing (Emekli-Alturfan et al., 2009). They also showed low F (0.02–0.04 mg/L) from herbal tea infusions, similar to our study (0.06–0.69 mg/L). This could be because these herbal teas had little *C. sinensis* leaves, which is the main contributor of F (Fung et al., 1999). It was reported that a substantial amount of F is released during tea infusion, with ~95% of F is available to consumers. Kalayci and Somer (2003) reported >96% F soluble in tea infusion. In black teas, high level of F at 6.1–7.3 mg/L was reported (Wong et al., 2003). In a study on Turkey and Iran teas, F contents were 35–289 mg/kg in tea samples compared to 0.64–3.92 mg/L in infusion (Hudaykuliev et al., 2005; Mahvi et al., 2006). In 56 Chinese puerh tea, F contents were 80.2–1526 mg/kg vs. 0.5 mg/L in infusion (Lv et al., 2013).

3.2. Daily fluoride intake in children and adults

As a popular beverage, the top three tea consuming countries include Turkey, Ireland and UK, where average annual tea consumption per person accounted for 6.9, 4.8 and 4.3 pounds (Ferdman, 2014). Tea is also a primary source of F, with low quantity being beneficial to health. However, excessive F intake over a prolonged period may result in fluorosis, which is characterized by dental mottling and skeletal manifestations such as crippling deformities, osteoporosis, and osteosclerosis (WHO, 2006). Being a F accumulator, health risk of F associated with tea consumption has been reported in traditional tea drinking regions of the world. For example, fluorosis in people in Sichuan, China has been associated with drinking a large amount of black tea (Chao et al., 1995). Further, a case of fluorosis has also been reported in an American patient having a history of consuming instant tea (Whyte et al., 2005). Therefore, it is pertinent to evaluate the daily intake of F in tea drinkers to assess the associated health risks.

Considering a daily tea consumption of 0.075 L by children <15 years and 0.75 L by adults >35 years (Sofuoglu and Kavcar, 2008), we calculated the daily F intake from tea in both (Fig. S1 and Fig. 1). For both children (0.004–0.05 mg/d) and adults (0.04–0.5 mg/d), the minimum F intake per day was associated with the consumption of herbal tea. Higher F intakes were associated with black tea

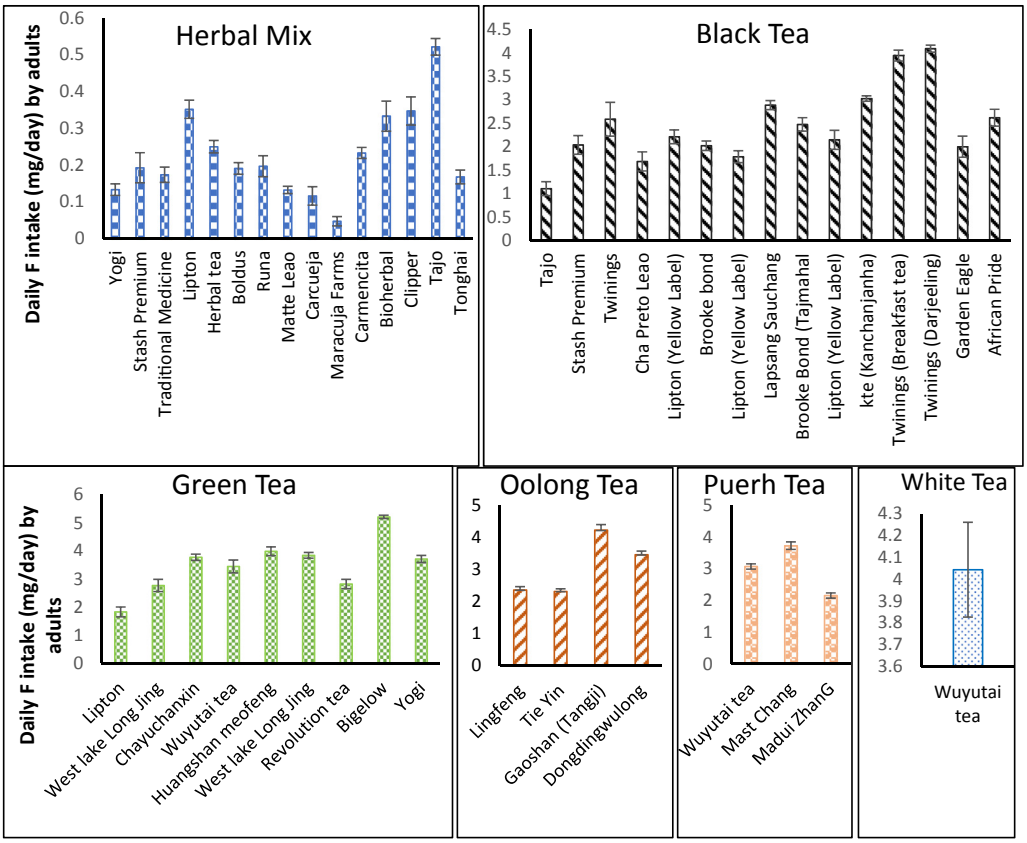

Fig. 1. Daily F (mg/d) intake by adults from traditional and herbal teas.

(0.11–0.41 mg/d in children and 1.1–4.1 mg/d in adults), green tea (0.18–0.52 and 1.8–5.2 mg/d), oolong tea (0.23–0.42 and 2.3–4.2 mg/d), puerh tea (0.21–0.30 and 2.1–3.0 mg/d) and white tea (0.4 and 4.0 mg/d) consumption.

Suggestions have been made with respect to the optimum F intake, ranging from 2 to 4 mg/d. For adults, the recommended safe daily intake from all sources is 1.5–4.0 mg and it is 1.5–2.5 mg for children (USEPA, 1987). We found that F levels from all teas were within the safe limits for both adults and children. However, we did not consider F from other sources, including drinking water and food, which would push the F load above the safe limit. Especially, in some areas of China (3.32 mg/L) and Tunisia (2.4 mg/L) where high F has been reported in water supply, the F risk associated with drinking tea may aggravate the problem (Pi et al., 2015; Guissouma et al., 2017). In our study, the highest daily F consumption from a green tea might contribute 5.2 mg F/d for adults. Cao et al. (2006) reported that an increased risk occurs for bone effect at F intake of 6 mg F/d whereas Whyte et al. (2005) considered 10 mg F/d as the threshold level for preclinical skeletal fluorosis.

3.3. Health risk assessment of F associated with drinking tea

Over the years, the health benefits of different teas have been studied (Yao et al., 2004). However, the detrimental effects of excessive F from drinking tea on human health cannot be ignored while considering its health benefits. Estimation of the chronic daily intake (CDI) of F in tea consumers can help to assess the risk involved (Sofuoglu and Kavcar, 2008). DRI (1997) has set the adequate intake of F from all sources in humans, including water, beverages and dietary allowances, at 0.05 mg/d/kg bw. Fig. S2 and Fig. 2 show the CDI values for F from teas in children and adults. For children, for all teas, the CDI values were below 0.05 mg/d/kg bw (Fig. S2). Similarly, for adults, none of the herbal teas had CDI above the stipulated levels. However, 2 black teas, 5 green teas, 1 oolong, 1 puerh and 1 white tea showed CDI values more than the stipulated value (Fig. 2). Strangely, out of 10 teas with high CDI values, 5 were green teas. Green tea has been used as a prophylactic drink by Chinese since ancient times due to its health benefits (Jain et al., 2013). However, occurrence of F can be a concern, which may reduce its marketability.

Normally, children below 8 years are susceptible to dental fluorosis during calcification process of teeth (Chouhan and Flora, 2008). Hence, hazard quotient (HQ) was estimated in children, keeping the risks of dental fluorosis in mind. Dental fluorosis, in its mild form, is characterized by white opaque areas covering 50% of a given tooth; in its severe form, dental fluorosis is characterized by brown to black stains and pitting (Chouhan and Flora, 2008). A value of 0.06 mg F/kg/d is the RfD in children for dental fluorosis and 0.12 mg F/kg/d is the RfD in adults for skeletal fluorosis (USEPA, 1987).

Fig. S3 and Fig. 3 show the HQ values for dental fluorosis in children and skeletal fluorosis in adults associated with drinking tea. HQ > 1 indicates health hazard associated with F from tea consumption. However, none of the teas posed an immediate hazard for dental fluorosis in children or skeletal fluorosis in adults. For children, HQ values were 0.003–0.04 in herbal teas, with the highest HQ associated with black, green, oolong, puerh and white tea at 0.31–0.43 (Fig. S3). Similarly, for adults, the highest HQ values were associated with black, green, oolong, puerh and white tea at 0.44–0.62 (Fig. 3). Although F from tea may not pose imminent threat to tea drinkers, exposure from other sources should also be considered, which add to daily F intake. However, we can conclude

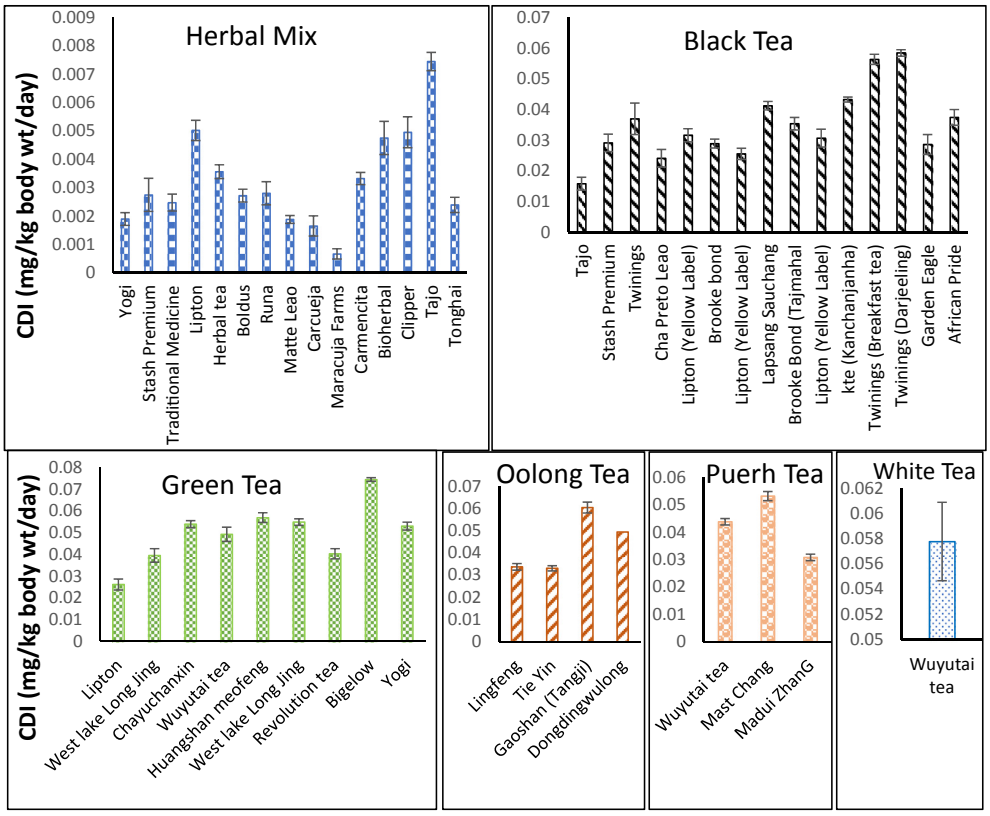

Fig. 2. Chronic daily intake (CDI) of F by adults from traditional and herbal teas.

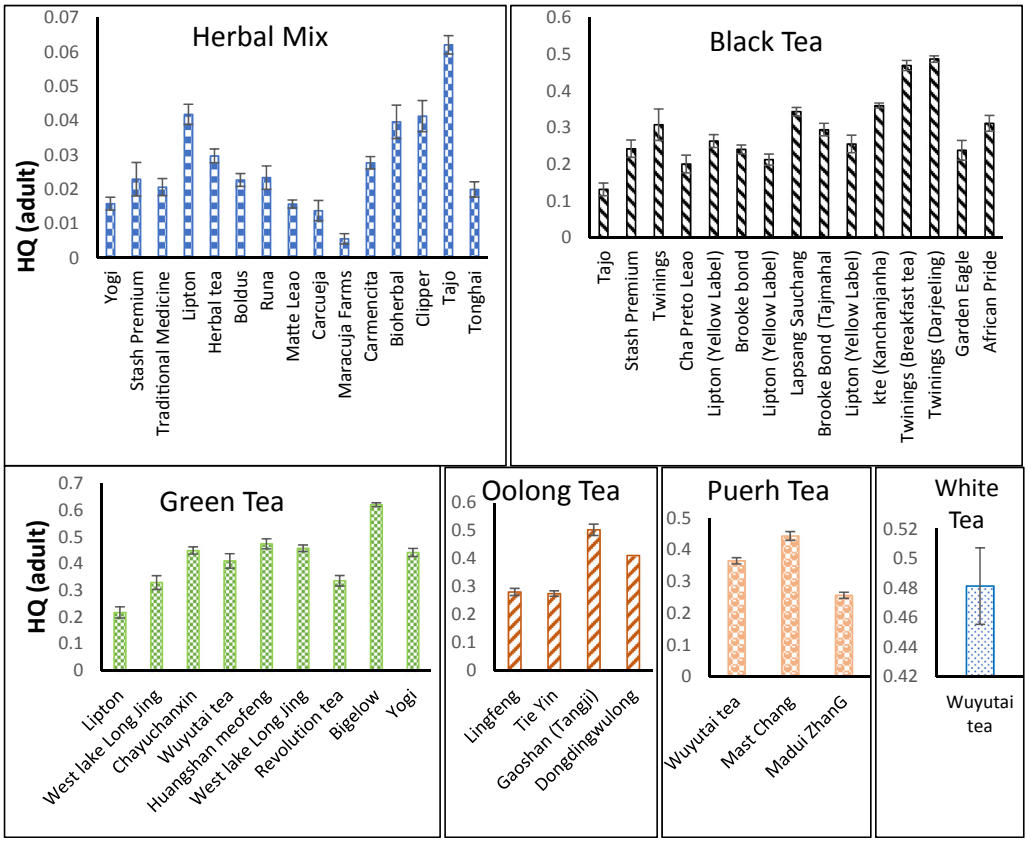

Fig. 3. Hazard quotient (HQ) for skeletal fluorosis in adults from traditional and herbal teas.

that heavy drinkers of traditional tea may be exposed to excess F, causing dental fluorosis in children and skeletal fluorosis in adults.

4. Conclusions

In this study, we assessed the health risk of F associated with the consumption of 47 traditional and herbal teas. The level of F was the least in herbal teas (33–102 mg/kg) and their infusion (0.06–0.69 mg/L) compared to traditional teas (296–1112 mg/kg) and their infusions (1.47–6.9 mg/L). During tea infusion, 6–96% and 18–99% of the total F in herbal and traditional teas were released into solution. The CDI values exceeded the stipulated value of 0.05 mg/d/kg bw in 10 teas, including 5 green teas, but none of the herbal teas exceeded the CDI value. Considering the HQ, drinking teas posed no imminent health hazards to both adults and children. However, since we did not consider F from food and drink sources, health risks from tea consumption cannot be ruled out. Hence, it can be predicted that long-term consumption of copious quantities of traditional tea might increase the chances of fluorosis in the consumers.

Acknowledgment

This study is supported in part by University of Florida. SD is thankful to the Department of Biotechnology (DBT), India for the award of DBT-Overseas Associateship (N.E) and Assam University for granting deputation to work in University of Florida.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2017.08.083.

References

Atoui, A.K., Mansouri, A., Boskou, G., Kefalas, P., 2005. Tea and herbal infusions: their antioxidant activity and phenolic profile. Food Chem. 89, 27–36.
Ayoob, S., Gupta, A.K., 2006. Fluoride in drinking water a review on the status and stress effects. Crit. Rev. Environ. Sci. Technol. 36, 433–487.
Barbier, O., Arreola-Mendoza, L., Del Razo, L.M., 2010. Molecular mechanisms of fluoride toxicity. Chemico-Biol. Interact. 188, 319–333.
Cao, J., Bai, X., Zhao, Y., Liu, J., Zhou, D., Fang, S., Jia, M., Wu, J., 1996. The relationship of fluorosis and brick tea drinking in Chinese Tibetans. Environ. Health Perspect. 104, 1340–1343.
Cao, J., Zhao, Y., Li, Y., Deng, H.J., Yi, J., Liu, W.J., 2006. Fluoride levels in various black tea commodities: measurement and safety evaluation. Food Chem. Toxicol. 44, 1131–1137.
Chao, J., Zhao, Y., Liu, J.W., Bai, X.S., Zhou, D.Y., Fang, S.L., Jie, M., Wu, J.S., 1995. Research on fluorosis caused by drinking brick tea in Sichuan Province. Chin. J. Tea Sci. 35–38.
Chouhan, S., Flora, S.J.S., 2008. Effects of fluoride on the tissue oxidative stress and apoptosis in rats: biochemical assays supported by IR spectroscopy data. Toxicology 254, 61–67.
Das, S., de Oliveira, L.M., da Silva, E., Ma, L.Q., 2017. Arsenate and fluoride enhanced each other's uptake in As-sensitive plant *Pteris ensiformis*. Chemosphere 180, 448–454.
Standing Committee on the Scientific Evaluation of the Dietary Reference Intakes, 1997. Dietary Reference Intakes (DRI) for Calcium, Phosphorus, Magnesium, Vitamin D, and Fluoride. National Academy Press, Washington, DC.
Emekli-Alturfan, E., Yarat, A., Akyuz, S., 2009. Fluoride levels in various black tea, herbal and fruit infusions consumed in Turkey. Food Chem. Toxicol. 47, 1495–1498.
Ferdman, R.A., 2014. Map: The Countries that Drink the most Tea. The Atlantic. Published on 21st January, available at: https://www.theatlantic.com/international/archive/2014/01/mapthecountriesthatdrinkthemosttea/283231/.
Fung, K.F., Zhang, Z.Q., Wong, J.W.C., Wong, M.H., 1999. Fluoride contents in tea and soil from tea plantations and the release of fluoride into tea liquor during infusion. Environ. Pollut. 104, 197–205.
Gramza-Michałowska, A., Kobus-Cisowska, J., Kmiecik, D., Korczak, J., Helak, B., Dziedzic, K., Górecka, D., 2016. Antioxidative potential, nutritional value and sensory profiles of confectionery fortified with green and yellow tea leaves (*Camellia sinensis*). Food Chem. 211, 448–454.
Guissouma, W., Hakami, O., Al-Rajab, A.J., Tarhouni, J., 2017. Risk assessment of fluoride exposure in drinking water of Tunisia. Chemosphere 177, 102–108.
Hudaykuliev, Y., Tastekin, M., Poyrazoglu, E.S., Baspinar, E., Velioglu, Y.S., 2005. Variables affecting fluoride in Turkish black tea. Fluoride 38, 38–43.
Jain, A., Manghani, C., Kohli, S., Nigam, D., Rani, V., 2013. Tea and human health: the dark shadows. Toxicol. Lett. 220, 82–87.
Kalayci, S., Somer, G., 2003. Factors affecting the extraction of fluoride from tea: application to three samples. Fluoride 36, 267–270.
Kalny, P., Fijałek, Z., Daszczuk, A., Ostapczuk, P., 2007. Determination of selected microelements in Polish herbs and their infusions. Sci. Total Environ. 381, 99–104.
Lv, H.P., Lin, Z., Tan, J.F., Guo, L., 2013. Contents of fluoride, lead, copper, chromium, arsenic and cadmium in Chinese Puerh tea. Food Res. Int. 53, 938–944.
Mahvi, A.H., Zazoli, M.A., Younecian, M., Esfandiari, Y., 2006. Fluoride content of Iranian black tea and tea liquor. Fluoride 39, 266–268.
Malde, M.K., Bjorvatn, K., Julshamn, K., 2001. Determination of Fluorine in food by the use of alkali fusion and fluoride ion-selective electrode. Food Chem. 73, 373–379.
Malinowska, E., Inkielewicz, I., Czarnowski, W., Szefer, P., 2008. Assessment of fluoride concentration and daily intake by human from tea and herbal infusions. Food Chem. Toxicol. 46, 1055–1061.
Martín-Domingo, M.C., Pla, A., Hernández, A.F., Olmedo, P., Navas-Acien, A., Lozano-Paniagua, D., Gil, F., 2017. Determination of metalloid, metallic and mineral elements in herbal teas. Risk assessment for the consumers. J. Food Compos. Anal. 60, 81–89.
Mitchell, W.J., Suggs, J.C., Bergman, F.J., 1977. Collaborative Study of EPA Method 13A and Method 13B. Publication No. EPAe300/4e77e050. U.S. Environmental Protection Agency, Research Triangle Park, NC.
Pi, K., Wang, Y., Xie, X., Su, C., Ma, T., Li, J., Liu, Y., 2015. Hydrogeochemistry of co-occurring geogenic arsenic, fluoride and iodine in groundwater at Datong Basin, northern China. J. Hazard. Mater. 300, 652–661.
Quock, R.L., Gao, J.X., Chan, J.T., 2012. Tea fluoride concentration and the pediatric patient. Food Chem. 130, 615–617.
Ruan, J., Ma, L., Shi, Y., Han, W., 2004. The impact of pH and calcium on the uptake of fluoride by tea plants (*Camellia sinensis* L). Ann. Bot. 93, 97–105.
Salahinejad, M., Aflaki, F., 2010. Toxic and essential mineral elements content of black tea leaves and their tea infusions consumed in Iran. Biol. Trace Elem. Res. 134, 109–117.
Sofuoglu, S.C., Kavcar, P., 2008. An exposure and risk assessment for fluoride and trace metals in black tea. J. Hazard. Mater. 158, 392–400.
USEPA, 1987. Fluorine (Soluble Fluoride); CASRN 7782-41-4 (IRIS Chemical Assessment Summary, Washington, DC).
USEPA, 1992. Guidelines for Exposure Assessment, EPA/600/Z-92/001. Risk Assessment Forum, Washington, DC.
USEPA, 1999. Guidelines for Carcinogen Risk Assessment, NCEA-f-0644. Review Draft. Risk Assessment Forum, Washington, DC.
World Health Organization (WHO), 2006. In: Bailey, K., Chilton, J., Dahi, E., Lennon, M., Jackson, P., Fawell, J. (Eds.), Fluoride in Drinking-water. WHO Press, Switzerland.
World Health Organization (WHO), 2011. Guideline for Drinking-water Quality, fourth ed. World Health Organization, Geneva, Switzerland.
Whyte, M.P., Essmyer, K., Gannon, F.H., Reinus, W.R., 2005. Skeletal fluorosis and instant tea. Am. J. Med. 118, 78–82.
Wong, M.H., Fung, K.F., Carr, H.P., 2003. Aluminum and fluoride contents of tea,with emphasis on brick tea and their health implications. Toxicol. Lett. 137, 111–120.
Yang, C.S., Landau, J.M., 2000. Effects of tea consumption on nutrition and health. J. Nutr. 130, 2409–2412.
Yao, L.H., Jiang, Y.M., Shi, J., Tomas-Barberan, F., Datta, A., Singanusong, R., Chen, S.S., 2004. Flavonoids in food and their health benefits. Plant Foods Hum. Nutr. 59, 113–122.