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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

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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.
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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

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

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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