Dietary risk assessment of fluoride, lead, chromium, and cadmium through consumption of Tieguanyin tea and white tea

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Abstract

Food and beverage consumption is the most possible route of human exposure to undesirable elements. Tieguanyin tea and white tea are widely-consumed beverage, which may be contaminated by fluoride (F), lead (Pb), chromium (Cr), and cadmium (Cd). For this study, the contamination levels and dietary exposure risks of these elements were studied from 72 Tieguanyin tea samples and 40 white tea samples from Fujian Province, China. The average concentrations of these elements decreased in order from F, Pb, Cr to Cd in Tieguanyin tea, while white tea in order from F, Cr, Pb to Cd. Metal-to-metal correlation indicated a weak positive correlation between Pb/Cd pairs (Tieguanyin tea) and Pb/Cr pairs (white tea). A weak negative correlation pair was found for Cr/Cd in Tieguanyin tea. The hazard index levels were below one, suggesting that there was no significant non-carcinogenic health risk to tea drinking consumers under the current dietary intake. The total cancer risk values of Pb, Cd, and Cr via the consumption of Tieguanyin tea and white tea were 4.01×10^{-5} and 7.64×10^{-6}, fell into the acceptable range of $10^{-6} \sim 10^{-4}$. It demonstrated that the cancer risk for consumers via drinking Tieguanyin tea and white tea was acceptable.

Keywords: tea; fluoride; heavy metals; health risk; cancer risk.

Practical Application: Food and beverage consumption is the most possible route of human exposure to undesirable elements. Due to potential pollution in plantation and processing procedures, tea may be contaminated with fluoride, lead, chromium, and cadmium. This situation may cause the emergence of many serious problems in terms of human health such as anemia, cancer, and miscarriages. Therefore, monitoring and health risk assessment of tea are in an urge.

1 Introduction

As the second most popular non-alcoholic beverage around the world, after water, tea (*Camellia sinensis*) is an important agricultural product for export in China (Yi et al., 2013; Zhang et al., 2017a). Depending on the fermentation process, teas are classified into three main categories, i.e. unfermented green tea, semi-fermented oolong tea and fermented black tea (Malinowska et al., 2008; Zhao et al., 2011; Yen et al., 2013; Castañeda-Saucedo et al., 2020). Compared to all other teas, white tea is produced in a special way, including only withering and drying processes without enzyme deactivation or fermentation (Chen et al., 2019; Dai et al., 2017; Ning et al., 2016). Because this simplest process, the high concentrations of catechins, amino acids, and other constituents are retained in white tea (Chen et al., 2019; Tian & Huang, 2019; Zhang et al., 2019; Ning et al., 2016). For another tea (Tieguanyin tea), though with high degree of fermentation, it is famous for the unique and elegant floral aroma, and ripe fruity flavor (Zhou et al., 2019; Xu et al., 2018). Both Tieguanyin tea and white tea are famous around the world and are mainly produced in Fujian Province (Li et al., 2014; Ning et al., 2016).

At present, the consumers are concerned about the chemical contaminants in food and beverages due to their potential toxicity to humans (Lv et al., 2013; Jiang et al., 2019). This issue has also received more attention concerning tea as the use of agro chemicals has increased over the past decades (Karak & Bhagat,

2010; Pehrsson et al., 2011; Yaqub et al., 2018). Fluoride, one of the most chemically active elements occurring in nature, exhibits both beneficial and toxic effects on human health (Janiszewska & Balcerzak, 2013; Chan et al., 2013). The tea plant (*Camellia sinensis*) takes up fluoride from soil and accumulates fluoride in its leaves (Fung et al., 1999). Sha & Zheng (1994) reported that 98% of fluoride in the whole tea plant were accumulated in tea leaves. Due to tea contains a very high quantity of fluoride and the transfer rate of fluoride during tea infusion is nearly 100% (about 94.9%) (Fung et al., 1999), tea is considered as the main natural source of fluoride intake (World Health Organization, 1984). And it has long been considered beneficial for the prevention of dental problems (Goenka et al., 2013). However, several studies have reported that the fluoride intake is one of the greatest potential health risks for tea consumers when its concentration exceeds a critical range (Emekli-Alturfan et al., 2009; Chan et al., 2013; Lv et al., 2013). Many studies show excessive fluoride intake may increase the incidence of dental fluorosis and skeletal fluorosis (Cao et al., 2005; Baskaradoss et al., 2008; Malinowska et al., 2008). There is even literature showing that exposure to fluoride may be a possible cause of cancer (Takahashi et al., 2001; Chan et al., 2013). The presence of toxic heavy metals (e.g., lead, chromium, cadmium) are other major concerns. During the growing and processing procedures, tea may be contaminated with heavy metals (Cao et al., 2010; Ning et al.,

2011). Moreover, rainfall, dust, and fertilizers also may be the sources of the heavy metal content in tea (Nkansah et al., 2016). According to previous research, the presence of heavy metals in some tea infusions exceeds the maximum limits recommended by different organizations or countries, which could pose a variety of health risks such as anaemia, cancer, and miscarriages (Salahinejad & Aflaki, 2010; Karak & Bhagat, 2010; Lv et al., 2013; Roya & Ali, 2017).

In the last decade, considerable research has been conducted on the fluoride and heavy metal content of brick tea (Koblar et al., 2012; Lv et al., 2013; Zhang et al., 2017b), black tea (Koblar et al., 2012; Dambiec et al., 2013; Waugh et al., 2017; Zhang et al., 2017b), oolong tea (Koblar et al., 2012; Zhang et al., 2017b), and green tea (Koblar et al., 2012; Zhang et al., 2017b). However, little data on the fluoride and heavy metal content in Tieguanyin tea and white tea are available, and information about their health risk assessment can thus not be found. Therefore, it is necessary to determine and analyze the magnitude of contamination by these elements in Tieguanyin tea and white tea from their major production areas. And risk assessment is needed to verify the current status of these undesirable elements.

The aims of this study were to (1) determine fluoride, lead, chromium, and cadmium concentrations in Tieguanyin tea and white tea from Fujian Province, China; (2) explore the correlation of the heavy metals with the Pearson correlation matrix; and (3) assess the potential risks fluoride and those heavy metals pose to consumer health.

2 Materials and methods

2.1 Samples collection and preparation

A total of 112 tea samples (500 g each), including 72 Tieguanyin tea samples and 40 white tea samples were randomly collected from the major producing counties [Anxi (24°50' -25°26' N; 117°36'-118°17' E), Hua'an (24°38' -25°12' N; 117°16'-117°42' E) and Fuding (26°52' -27°26' N; 119°55'-120°43' E)] (Figure 1) by the agricultural bureau in the counties involved. The sampling was done according to guideline in China (Standardization Administration of China, 2013). An amount of 200 g of each tea sample was ground and passed through a 200 μm polyethylene sieve. All the powdered tea samples were stored in separate polyethylene container at 4 °C until analysis.

2.2 Chemicals and reagents

All chemical products used were of analytical reagent grade unless otherwise stated. Ultrapure water was obtained using a Direct-Q3 UV water system (18.25 MΩ cm^{-1}, Millipore, Bedford, MA, USA) for all dilutions and blanks. Hydrochloric acid (HCl, 37%, Analytical reagent) and nitric acid (HNO$_3$, 65%, guaranteed reagent) were provided from Xilong Chemical Co., Ltd. (Shantou, Guangdong, China). The calibration curves were prepared using dilutions of F, Pb, Cr, Cd standard solution (Guobiao Testing & Certification Co., Ltd., Beijing, China). Tea certified reference material (GBW10016, Institute of Geophysical and Geochemical Exploration, China) was used for quality assurance.
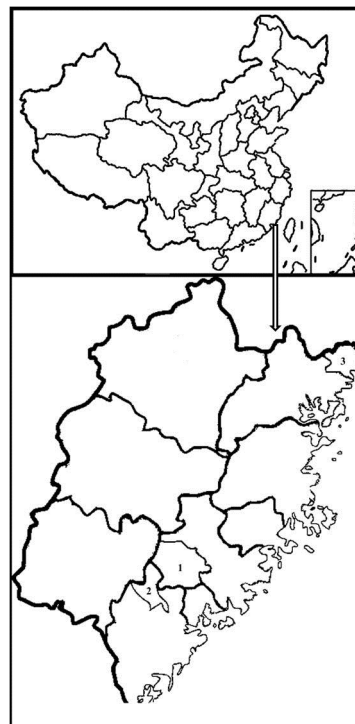

Figure 1. Production of areas of Tieguanyin tea and White tea in Fujian province of China: (1) Anxi; (2) Hua'an; (3) Fuding.

2.3 Determination of fluoride

The total fluoride content in the tea samples was analyzed according to the standard method for analysis of fluoride in food (Standardization Administration of China, 2003). Briefly, 1.000 g tea sample was extracted by10 mL of 0.2 M HCl for 1 h with occasional gentle shaking. The extract of tea samples or F standard solution was added to 25 mL total ionic strength adjusting buffer, then de-ionized water was added up to 50 mL. A fluoride ion-selective electrode (INESA Scientific Instrument Co., Ltd., Shanghai, China) was used to measure the fluoride content. Prior to sampling and at the end of group of five samples, the electrode was re-checked for accuracy. The reliability of the method was certified by standard reference material of tea (GBW10016). The agreement between results and the certified values was satisfactory, the mean recovery was 94% with a standard deviation (SD) 8%. Three replicates of each tea sample undergoing the same procedures were performed. Data are expressed on a dry weight (DW) basis.

2.4 Determination of lead, chromium, and cadmium

Inductively coupled plasma mass spectrometry (ICP-MS) (XSERIES 2, Thermo Fisher Scientific Co., Ltd., MA, USA) was used for determination of Pb, Cr, Cd according to previous research (Han et al., 2005; Lv et al., 2013). All glassware was kept in the 10% HNO$_3$ solution and rinsed with ultra-pure water, then air dried before use. A microwave digestion system (TOPEX, Preekem Scientific Instruments Co. Ltd., Shanghai, China) was applied for sample digestion. About 0.5 g of each tea sample was weighed and 5 mL of high purity HNO$_3$ was added. The mixture was left at room temperature for 15 min and then

placed in the microwave oven for 10min at a maximal pressure of 800 psi and 170°C temperature. After digestion and cooling, each solution was evaporated to near dryness and transferred into a 25 mL volumetric flask. The solution was topped up to the mark using de-ionized water and transferred into a PTFE bottle, ready for ICP-MS analysis. The mean recoveries for these 3 elements ranged from 90% to 115%, with a standard deviation (SD) lower than 10%. The limits of detection (LOD) of Pb, Cd, and Cr were 0.005, 0.001, and 0.05 mg/kg, respectively. The accuracy and ICP-MS instrument performance were evaluated by the tea certified reference material (GBW10016). A blank digestion solution was made for comparison. All samples were measured in triplicate. Data are expressed on a dry weight (DW) basis.

2.5 Statistical analyses

Statistical analysis of data was performed with SPSS (Statistical Package for Social Sciences) version 20 for Windows (SPSS Inc., Chicago, IL, USA). Results were expressed as the mean ± standard deviation (SD). Mean concentrations of each element in different origins or varieties were evaluated for significance by one-way analysis of variance (ANOVA). A statistically significant difference between different sets of data was defined as a value of $p<0.05$. The Pearson's correlation matrix was used to analyze the relationships between heavy metal concentrations. It is useful for indicating the pollutants' probable common source.

2.6 Health risk assessment

The potential health risk, including non-cancer health risk and target cancer risk via drinking tea infusion for adults, were assessed as reported by Castro-González et al. (2019) and Bortey-Sam et al. (2015). The non-cancer health risk was performed by calculating the target hazard quotient (THQ), by dividing the chronic daily intake (CDI) with the oral reference dose (RfD) (Equation 1, Equation 2).

$$CDI = (C \times D \times T) \div Bw \qquad (1)$$

where C is the average content ($\mu g\ kg^{-1}$) of a trace element in tea, D is the amount of tea consumed, T is the transfer rate of a trace element from made tea to tea infusion and Bw is the average body weight.

$$THQ = CDI / RfD \qquad (2)$$

where RfD values of the trace elements were obtained from the US Environmental Protection Agency (USEPA) or the Joint FAO/WHO Expert Committee of Food Additives (JECFA), which are given in Table 1. A hazard index (HI), sum of THQ (Equation 3), was obtained to determine the overall potential risk of Pb, Cd and Cr.

$$HI = \Sigma THQ \qquad (3)$$

If the HI value is above or equal to 1, this indicates an unacceptable health risk for human health (Khan et al., 2013; United States Environmental Protection Agency, 2016).

Table 1. Oral reference dose (RfD) values and slope factors (Sf) of the metals considered to be carcinogenic.

Trace elements	RfD ($\mu g\ kg^{-1} day^{-1}$)	Sf ($mg\ kg^{-1}\ day^{-1}$)
F	4000	-
Pb	3.57[a]	0.0085[b]
Cr	3	0.5
Cd	1	6.1

[a]Pb is set at 3.57 ug kg^{-1} day^{-1} according to provisional tolerable daily intake (PTDI) suggested by FAO/WHO, since the RfD for Pb in USEPA is not available. [b]Cd, Cr are the heavy metals considered to be carcinogenic and Pb is probably carcinogenic.

Target cancer risk (TR) was used to indicate carcinogenic risks from the consumption of tea. The model for estimating TR was shown as follows (Equation 4):

$$TR = CDI \times Sf \qquad (4)$$

where Sf is the slope factors of the heavy metals considered to be carcinogenic. The total cancer risk (TR_{total}) was calculated as (Equation 5):

$$TR_{total} = \Sigma TR \qquad (5)$$

3 Results and discussion

3.1 Contaminant levels of fluoride, lead, chromium, and cadmium in tea

The analytical results of 112 tea samples collected from different areas of Fujian Province are summarized in Table 2. Concentration of the elements analyzed were in the range 48-580 mg kg^{-1} for F, 0.18-2.00 mg kg^{-1} for Pb, 0.08-2.30 mg kg^{-1} for Cr, and 0.01-0.11 mg kg^{-1} for Cd, respectively. According to this data, F had the highest concentration, followed by Pb, Cr, and Cd.

Fluoride

Fluoride mean contents in tea samples collected from different areas were 190 mg kg^{-1}(Anxi), 188 mg kg^{-1} (Hua'an), 142 mg kg^{-1} (Fuding). The highest content of F was found in a Tieguanyin tea sample from Hua'an and the lowest one was found in a white tea sample from Fuding. The average fluoride concentration in white tea was significantly lower than that in Tieguanyin tea ($p<0.05$). However, no significant difference was observed between the average fluoride content in Tieguanyin tea from different production areas. A plausible explanation could be the different manufacturing procedure of the white tea in comparison to the Tieguanyin tea (Tan et al., 2017; Sanlier et al., 2018). Tieguanyin tea was often processed from one bud and four or five leaves, while only one bud and three leaves were picked for white tea, and their tea plant varieties were different (Sanlier et al., 2018; Xu et al., 2018). Simultaneously, Tieguanyin tea is semi-fermented and white tea is not fermented. Shu et al. (2003), Malinowska et al. (2008), and Janiszewska & Balcerzak (2013) revealed that the content of fluoride in tea depends on maturity and the fermentation step of processing. The fluoride content in black tea is generally higher than that of non-fermented

Table 2. Content of fluoride, lead, chromium and cadmium in tea from Fujian Province of China (mg kg^{-1}).

Tea type, Area	Tieguanyin, Anxi (n=40)		Tieguanyin, Hua'an (n=32)		White tea, Fuding (n=40)		Mean ± SD
Elements	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	
Fluoride	73-350	190 ± 78[b]	89-580	188 ± 101[b]	48-290	142 ± 61[a]	173 ± 83[ab]
Lead	0.31-2.00	0.91 ± 0.41[b]	0.40-1.28	0.81 ± 0.24[b]	0.18-1.69	0.65 ± 0.37[a]	0.79 ± 0.34[ab]
Chromium	0.11-1.38	0.58 ± 0.30[ab]	0.08-1.20	0.54 ± 0.27[a]	0.14-2.30	0.73 ± 0.52[b]	0.62 ± 0.39[ab]
Cadmium	0.01-0.11	0.04 ± 0.03[b]	0.01-0.05	0.02 ± 0.01[a]	0.01-0.07	0.03 ± 0.01[a]	0.03 ± 0.02[a]

[ab]Means with the different letters in row are significantly different ($p<0.05$). SD, standard deviation.

or semi-fermented teas (Shu et al., 2003; Malinowska et al., 2008). Zhang et al. (2017a) also showed that the average contents of fluoride in different types of teas decreased in order from brick tea, to black tea, oolong tea, and green tea. The brick tea produced in Sichuan Province contained high fluoride concentrations with a mean value 491.8 mg kg^{-1} (Cao et al., 1998). Furthermore, the difference in the mean fluoride concentration between Tieguanyin tea and white tea may be partly associated with soil conditions. It was reported that the plantations' soil conditions and the tea plant varieties should influence the fluoride content in tea (Shu et al., 2003; Lv et al., 2013).

Lead

Lead levels in tea samples in this study were lowest in a white tea sample collected from Fuding and highest in a Tieguanyin sample from Anxi. The average lead content of tea from different production areas was 0.91 mg kg^{-1} (Tieguanyin tea, Anxi), 0.81 mg kg^{-1} (Tieguanyin tea, Hua'an), and 0.65 mg kg^{-1} (white tea, Fuding), respectively. The significantly higher mean lead content for Tieguanyin tea than for that of white tea can be explained by the different types of leaves which are picked for them. With the different tea plant varieties, Tieguanyin tea was processed from one bud and four or five leaves, while white tea was processed from one bud and three leaves (Sanlier et al., 2018; Xu et al., 2018). Furthermore, older tea leaves tend to contain higher concentrations of lead than younger leaves (Natesan & Ranganathan. 1990). The previous studies found the lead content in different tea types increase in order from green tea, to black tea, and oolong tea (Han et al., 2006, 2014). Moreover, the processing procedure and storage (e.g. containers) could be other probable sources of the lead content in tea.

Chromium

The tea samples from Anxi, Hua'an, and Datian showed average Cr Concentrations of 0.58 mg kg^{-1}, 0.54 mg kg^{-1}, and 0.73 mg kg^{-1}, respectively. The highest and the lowest content of Cr were observed in a Tieguanyin tea sample from Anxi and white tea sample from Fuding. The Cr content of white tea was higher than that of Tieguanyin tea from Hua'an ($p< 0.05$). However, comparison of Cr content in Anxi and Fuding tea samples showed no significant differences ($p> 0.05$). In a survey done on a total of 801 tea samples in China, the highest mean content of Cr was found in black tea (Han et al., 2005). Additionally, significant differences were observed between green tea and black tea, as well as black tea and oolong tea (Han et al., 2005). Seenivasan et al. (2008) reported that Cr content in tea was mainly correlated with the sharpening of crush, tear, and

curl rollers used for manufacturing. It has also been reported that Cr contamination of plants depends on Cr speciation in soil (Gardea-Torresdey et al., 2004). This could be partly explained by the difference in Cr concentration in the tea from different production areas.

Cadmium

The Cd content in Anxi tea samples varied between 0.01 mg kg^{-1} and 0.11 mg kg^{-1} with a mean of 0.04 mg kg^{-1}, which was significantly higher than that in tea from Hua'an or Fuding ($p<0.05$). There was no significant difference ($p>0.05$) between the concentration of Cd in tea samples from Hua'an and Fuding. Franklin et al. (2005) indicated that cadmium in tea was mainly absorbed from zinc fertilizers and phosphate fertilizers. And the mean concentration of Cd in this study was lower than Cd contents in black tea (Ashraf & Mian, 2008; Nkansah et al., 2016) and green tea (Han et al., 2005; Nkansah et al., 2016). However, the contaminated level of Cd in oolong tea (Han et al., 2005), Pu'er tea (Ning et al., 2011) and green tea (Nookabkaew et al., 2006) were found to be comparable to the present study.

3.2 Pearson's correlation analysis for lead, chromium, and cadmium

The Pearson's correlation analysis is a measure of the linear correlation between two variables and shows the results in a matrix form. Pearson's correlation is helpful for showing the direction and strength of the association among variables. The values are between +1 and -1, where 1 is total positive linear correlation, 0 is no linear correlation, and -1 is total negative linear correlation. Nkansah et al. (2016) reported that the heavy metals in tea may have a probable common source if the pair of metals has a close relationship. Metal-to-metal correlation coefficient matrix for tea samples is shown in Table 3. The significantly weak positive correlation was found for Pb/Cd pairs (r=0.392, $p<0.01$) in Tieguanyin tea and Pb/Cr pairs (r=0.350, $p<0.05$) in white tea, indicating that these heavy metals may have a common pollution source. Cr concentration in Tieguanyin tea showed a relatively weak negative correlation with Cd concentration (r=-0.297, $p<0.05$). The values of r for Pb/Cr in Tieguanyin tea were not at significant levels ($p>0.05$). And Cd concentration of white tea also showed no significant correlation with Pb or Cr concentration ($p>0.05$). A similar study of Nkansah et al. (2016) showed Cd/As, and Pb/As in tea from Ghana of may have a probable common source. In general, the mechanism of heavy metal enrichment in tea is complex and is related to soil physicochemical properties of the tea plantations, the age of tea trees, the elements' properties and speciation (Jin et al.,

2005; Yemane et al., 2008). Furthermore, tea could be also contaminated by heavy metals during the processing procedure (Qin & Chen, 2007). Hence, research should be conducted in the future to find out the clear sources of these trace elements in Tieguanyin tea and white tea.

3.3 Health risk assessment

Non-cancer health risk

The health risk assessment was only performed for adults due to children rarely having the habit of drinking tea. As with other beverage crops, water infusion is the main form of tea consumption. Therefore, the transfer rate of trace elements from processed tea leaves to tea infusion needs to be considered when calculating the actual risk of them on human health. In the present study, the transfer rates of F, Pb, Cr, and Cd from made tea to tea infusion were quoted from the previous reports (Table 4).

The CDI values of trace elements through the consumption of Tieguanyin tea or white tea are presented in Table 4. The CDI values of trace elements in Tieguanyin tea decrease in order from F to Pb, Cr, and Cd, while in white tea in order from F to Cr, Pb, and Cd. The THQ values of each trace element were below one, indicating that the daily intake of each trace element content in Tieguanyin tea or white tea has no significant potential health risk to a normal adult, which is similar to many other teas assessed by former studies (Shen & Chen, 2008; Cao et al., 2010; Zhang et al., 2018). The trend of THQ values of individual trace elements via the consumption of Tieguanyin tea and white tea decreased in the order of Pb, Cr, F, Cd, and F, Cr, Pb, Cd, respectively. As shown in Table 4, the HI value was calculated to assess the combined non-carcinogenic effects of F, Pb, Cr, and Cd. The HI value of 4.53×10^{-2} was found for Tieguanyin tea,

and 1.31×10^{-2} for white tea. Both were far below one, indicating that there was no significant non-carcinogenic health risk to tea consumers (Fu et al., 2014; Nkansah et al., 2016).

Target carcinogenic risk of Pb/Cr/Cd

Aside from non-carcinogenic health risks, some heavy metal elements, such as Cr and Cd, are also carcinogenic. Pb is probably carcinogenic (Castro-González et al., 2019). The cancer slope factors of Cr, Cd, and Pb are shown in Table 1. The carcinogenic risk as a result of the consumption of Tieguanyin tea in this study, based on the individual effects of Pb, Cr, Cd, showed that Cr had the highest target cancer risk (TR) value of 2.23×10^{-5}, followed by Cd and the least by Pb (Figure 2). Meanwhile, the carcinogenic risk of individual effects of heavy metals in white tea was in the order of Cr > Pb > Cd. The total target carcinogenic risk (TR_{total}) of Pb, Cr and Cd via the consumption of Tieguanyin tea and white tea are 4.01×10^{-5} and 7.64×10^{-6}, which fall into the acceptable range of $10^{-6} \sim 10^{-4}$ as given by the United States Environmental Protection Agency (2002), demonstrating that the cancer risk of tea consumption was acceptable (Li et al., 2013; Bamuwamye et al., 2015).

In fact, several other uncertain factors may also affect chronic trace elements intake. For example, there are many other exposure ways on health risk, such as food, skin contact and other harmful elements (e.g. arsenic, aluminum, and mercury).
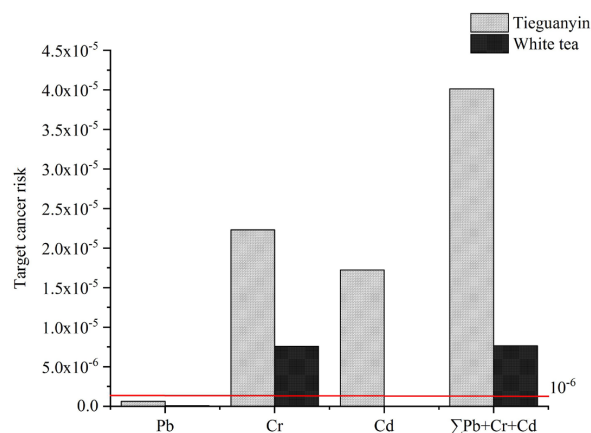
Table 3. Pearson correlation coefficients among metal concentrations ($r=95\%$).

	Tieguanyin			White tea		
	Pb	Cr	Cd	Pb	Cr	Cd
Pb	1.000	0.173	0.392**	1.000	0.350*	-0.150
Cr		1.000	-0.297*		1.000	-0.276
Cd			1.000			1.000

*Correlation is significant at the 0.05 level (two-tailed); **Correlation is significant at the 0.01 level (two-tailed).

Figure 2. Target cancer risk and Total target cancer risk index in consumers due to consumption of tea.

Table 4. Transfer rate, average content (C), chronic daily intake (CDI) and THQ of 4 elements in tea from Fujian Province of China.

	Tieguanyin				White tea			
	Fluoride	Lead	Chromium	Cadmium	Fluoride	Lead	Chromium	Cadmium
Transfer rate (%)[a]	94	50.1	47.5	52.7	100	7.1	12.4	ND
C (mg kg^{-1})	189	0.86	0.56	0.03	142	0.65	0.73	0.03
CDI[b] (µg kg^{-1}bw day^{-1})	29.68	7.19×10^{-2}	4.46×10^{-2}	2.82×10^{-3}	23.68	7.73×10^{-3}	1.52×10^{-2}	-
THQ	7.42×10^{-3}	2.02×10^{-2}	1.49×10^{-2}	2.82×10^{-3}	5.92×10^{-3}	2.17×10^{-3}	5.05×10^{-3}	-
HI	4.53×10^{-2}				1.31×10^{-2}			

[a]The transfer rates of F, Pb, Cr, and Cd from made tea to tea infusion were quoted from the reports of Shokrzadeh et al. (2008), Shen & Chen (2008) and Koblar et al. (2012). The transfer rates of these four elements for white tea are not available, so the value for green tea was used; [b]According to the report of Chen et al. (2015)[66], daily intake of Tieguanyin tea is 10 g and the adult body weight is 60 kg. ND, not detected.

And the health risk may increase due to the accumulation of heavy metal elements in the organism. Rasmussen et al. (2001) reported that Pb remains in the bones for decades. Furthermore, the occurrence forms and valence of heavy metal elements were not considered, so that the evaluation results, calculated with the total exposure dose, were overestimated. Thus, some deviations between the final health risk value and real value remain. As a conclusion, because the health risk through the consumption of Tieguanyin tea or white tea is low, the uncertainties that cause overestimation will not have a big influence on the assessment results.

4 Conclusion

This study assessed the health risk of F, Pb, Cr, and Cd in Tieguanyin tea and white tea from Fujian province of China. Significant differences were found in the content of these trace elements in tea from the different production areas. The average concentrations of these four elements in Tieguanyin tea were statistically significantly different compared with white tea ($p<0.05$). However, no significant differences were observed in the contaminated levels of the studied elements except Cd in Tieguanyin tea between Anxi and Hua'an. The significantly weak positive correlation was found for Pb/Cd pairs in Tieguanyin tea and Pb/Cd pairs in white tea. Cr concentration in Tieguanyin tea showed a relatively weak negative correlation with Cd concentration. The HI value in tea was below one, indicating the trace elements (F, Pb, Cr, and Cd) in Tieguanyin tea or white tea carry no non-carcinogenic health risk to tea consumers. The target cancer risk from exposure to these heavy metals by consuming tea was acceptable as the total target cancer risk (TR_{total}) was lower than the US-EPA management level of 10^{-4}. These results may provide a reference for tea stakeholders, including risk managers, tea industry, and consumers.

Acknowledgements

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