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Concentrations, leachability, and health risks of mercury in green tea from major production areas in China

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ABSTRACT

Green tea has many health benefits and is the most consumed type in China. However, the heavy metals and contaminants in tea can also pose a great risk to human health. In this study, mercury (Hg) concentration in green tea collected from 11 provinces in China was examined. The leaching characteristics of Hg during brewing and the associated exposure to drinkers were also evaluated. Results indicated a low potential of Hg accumulation in green tea. The Hg content of green tea from Wanshan District, Guizhou Province—which has the largest Hg mine in China and is severely contaminated by Hg—could be limited by controlling the harvest time of tea leaves. The average Hg content of green tea from 43 tea production sites in China was only 6.3 ± 6.4 μg/kg dry weight. The brewing experiments of green tea showed that the leaching ratio of Hg was 22.61 ± 7.58% for 40 min of a single brew, and increased to 32.83 ± 12.37% after four rounds (3 min/ round) of brewing. The leaching of Hg from tea leaves was significantly affected by leaching time, temperature, and solid-liquid ratio but not by water hardness. The risk of Hg exposure from green tea intake was found to be very low, with an average hazard quotient (HQ) value of only 1.82 ± 1.85% for a single brew in 40 min and 2.64 ± 2.68% after four rounds of brewing. However, in some highly contaminated areas, with HQ values as high as 43.12 ± 2.41%, green tea intake may still pose a high risk of Hg exposure, and this risk should not be ignored.

1. Introduction

Mercury (Hg) is a toxic heavy metal with a high propensity of bio-enrichment, persistence, and widespread contamination (Wang et al., 2021b). It exposure in human beings is associated with numerous toxic effects on the immune, digestive and nervous systems and on kidneys, lungs, eyes, and skin (Natasha et al., 2020). Even low doses of Hg exposure may have adverse effects on human health (Davidson et al., 2004; Feng et al., 2020; Gustin et al., 2020). Food and beverages consumption are the primary routes of Hg exposure in humans and have attracted increasing attention in recent years (Natasha et al., 2020; Wang et al., 2021a; Zhang et al., 2020).

The tea tree (*Camellia sinensis*, L.) has its origin in southwest China, where it has been cultivated for at least 2000 years (Chen et al., 2009). It is a nonalcoholic stimulating beverage with unique biological activities and health benefits (Klepacka et al., 2021; Xing et al., 2019) and is most widely consumed next to water (Xing et al., 2019). Despite the health benefits, residues (Yang et al., 2020) and contaminants (Abd El-Aty et al., 2014; Das et al., 2017; Peng et al., 2018; Yemane et al., 2008) in tea may pose health hazards to tea drinkers. There is a growing concern about Hg contamination in tea and infusions. Tea intake can increase the Hg content in the blood (Colapinto et al., 2016) and may accelerate the enterohepatic methylmercury (MeHg) cycle to contribute to a temporary bioamplification of MeHg in the bloodstream (Canuel et al., 2006).

China was the largest tea producer and exporter globally, accounting for 54% and 41% of global tea acreage and production, respectively, in 2016 (Zhang et al., 2018). However, Hg contamination was observed in areas with intense human activities and higher geological background in this country (Jiang et al., 2006; Li et al., 2009; Liu et al., 2021a), and this

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would lead to a significant increase in Hg content in crops and vegetables grown in these areas and consequently increases exposure risk to the local population (Aslam et al., 2020; Feng et al., 2008; Wang et al., 2020). In China, tea cultivation is conducted on a considerable scale. The difference in the geological background (Xu et al., 2022) and the complexity of the environment around tea growing areas may lead to severe contamination, especially of heavy metals, in some tea plantations (Li et al., 2021). Thus, the large scale of tea production and variety in environmental conditions across China make Hg exposure from tea intake an issue that needs urgent attention.

The risk of Hg exposure from tea intake is mainly influenced by two factors, one of which is Hg content in tea. Non-volatile metals in tea are mainly influenced by soil conditions, and their origin can even be traced by the elemental characteristics (Ana et al., 1998; Pilgrim et al., 2010). However, Hg contamination in tea may also be influenced by atmospheric Hg, its dry and wet deposition, and other factors (Woerndle et al., 2018; Wright et al., 2016), making the factors influencing Hg content in tea were very complex. The average Hg content in tea has been found to vary greatly (2–130 μg/kg) (Árvay et al., 2015; Chen et al., 2020; Falahi and Hedaiati, 2013; Jian et al., 2018; Karimi et al., 2008; Li et al., 2020; Ning et al., 2011; Nookabkaew et al., 2006; Pourramezani et al., 2019; Schulzki et al., 2017; Zhang et al., 2013, 2020) and the highest content was detected to be 410 μg/kg (Nookabkaew et al., 2006), much higher than the maximum permissible limit (100 μg/kg) recommended by the European Commission Regulation for certain contaminants in foodstuffs (2008) (Gasser et al., 2009) and by the European Pharmacopeia of herbal drugs (Martín-Domingo et al., 2017). It is also higher than the limit (300 μg/kg) defined by the Ministry of Agriculture Tea heavy metals limited standards in China (NY659, 2003). Although there have been many studies on Hg content in Chinese tea (Chen et al., 2020; Li et al., 2020; Zhang et al., 2013, 2020), there is still a lack of nationwide study on Hg content in tea, especially green tea. Based on the processing method, tea can be of several types, such as green tea, white tea, oolong tea, black tea, dark tea, and pu'erh tea (Ma et al., 2019). Green tea is a 'non-fermented' form of tea produced by drying and steaming the fresh leaves; it accounts for about 20% of the tea produced worldwide and is mainly consumed in Asian countries (Gruszecka-Kosowska and Mazur-Kajta, 2016). In China, green tea production accounted for 65.8–72.89% of the total tea production from 2000 to 2018 (Zhou and Wu, 2020). Green tea is normally produced from young shoots, usually picked within 30 days of sprouting. It has the lowest elemental content (Ma et al., 2019), with much lower Hg content in young than mature leaves (Árvay et al., 2015). Hence, the Hg content in green tea may be relatively low. Nevertheless, as a primary consumed tea type in China, Hg content in green tea needs to be evaluated.

Characteristically, Hg leaches out during the tea brewing process is another issue that deserves investigating. Tea is usually consumed in the form of tea infusion. Therefore, the Hg leaching ratio during tea infusion is critical for determining Hg exposure. While several factors affect the leaching ratio of chemicals during tea brewing (Chen et al., 2020; Lung et al., 2008), only a few studies have focused on the leaching property of Hg. Karimi et al. (2008) and Zhang et al. (2013) have shown that Hg has the highest release ratio (70% and 45%, respectively) during tea infusion compared to other elements including Pb, As, Cu, Cd, Cr, and Al. Hg in tea may pose a high risk of exposure to humans; therefore, it is important to study the effect of different tea brewing conditions on the leaching of Hg.

In this study, the Hg content in green tea samples collected from the main tea-producing areas in China was measured and compared with those from a tea garden located in the Wanshan mercury mining area (WMMA) in Guizhou Province, China, which is severely contaminated with Hg (Chang et al., 2020). The effects of tea brewing conditions such as temperature, duration, and water quality on Hg leaching were systematically investigated. The results obtained may lay the foundation for the risk assessment of Hg exposure to tea drinkers and the development of the tea industry in China.

2. Materials and methods

2.1. Site description and sampling

All the tea samples were collected from the main tea producing areas of China, including Fujian (FJ), Jiangxi (JX), Hubei (HB), Zhejiang (ZJ), Hunan (HUN), Shaanxi (SX), Guizhou (GZ), Sichuan (SC), Anhui (AH), Henan (HEN), and Yunnan (YN) provinces. As a control, the sample from TRCD (Tongrenchadian) from a tea garden in WMMA (sample site 43, Fig S1) was used, and the soil Hg content (157–1536 μg/kg) and bioaccumulation factors (average 4.9 ± 3.6%) of Hg from soils to green teas of those sites in Guizhou (including sample site 43) are mentioned in Table S2. All the sampling sites are indicated in Fig. 1. The total number of green tea samples was 129 (3 samples per site). All green tea samples were collected directly from the tea production factories to ensure that the tea was local and all samples were spring tea. The sampling period lasted from July 2020 to July 2021.

2.2. Mercury accumulation in tea leaves

Hg accumulation in tea leaves was monitored in the tea plantation garden located in sampling site No. 43. The sampling period lasted from March 3, 2021, to June 2, 2021. Tea leaves sprouting in the same period were marked with a thin red line, collected at different intervals, air dried, and then ground, and passed through a size 60 mesh to determine the Hg content.

2.3. Leaching and analysis

There are mainly two ways of tea brewing in China, one of which is single brewing (Yu et al., 2021). The specific steps are to place a certain amount of tea leaves in a container, add the right amount of hot water, and wait for the tea brew to cool down. The brewing time in this method usually lasts from a few minutes to dozens of minutes before drinking. The other way is multiple brewing (Lung et al., 2008), adapted from the ancient Chinese tea culture. The number of brewing is related to the tea type, usually within four times and each time lasting within 3 min. In this method, the tea leaves are placed in a teapot, hot water is added, and then quickly poured out into a teacup to cool down for drinking; the above process was repeated until the taste of the tea brew deteriorated. The specific leaching process of green tea was performed as described in a previous study (Lung et al., 2008), with a few modifications. Typically, the test tea sample was first divided into four portions of 2 g each and labeled as samples 1–4, respectively. The tea samples (1−4) were first placed in four glass containers, and then 150 mL of boiling water (97 ± 1 ℃) was added to each container. For single brewing, the samples 1–4 were brewed for 5, 10, 20, and 40 min, respectively, to evaluate the effect of time on the Hg leaching ratio. For the multiple brewing test, 2 g tea was brewed for 3 min and then filtered. The infusions of samples 1–4 and tea residues of sample 1 were collected. To the tea residues of samples 2–4 after filtration, 150 mL of boiling water was added, brewed for 3 min, and then filtered, and the infusions of samples 2–4 and tea residues of sample 2 were also collected. The above process was subsequently repeated to obtain 10 tea infusions and four tea residue samples. All the residues were then air-dried, and the infusions were stored for further testing. The effect of brewing conditions on Hg leaching was assessed by varying the brewing temperature (67, 77, 87, and 97 °C), solid-to-liquid ratio (1:50, 1:75, and 1:100) and water hardness. The effect of water hardness was simulated by adding different amounts of calcium carbonate (100, 200, and 300 mg/L) in water (Lung et al., 2008).

The Hg contents in tea and tea residues were analyzed using a Milestone Direct Mercury Analyzer (Model DMA-80, Milestone Italy), which we also used earlier to measure Hg content in plant samples (Wang et al., 2021a). The Hg concentration in tea infusions was measured by cold vapor atomic fluorescence spectroscopy (Model 2500,

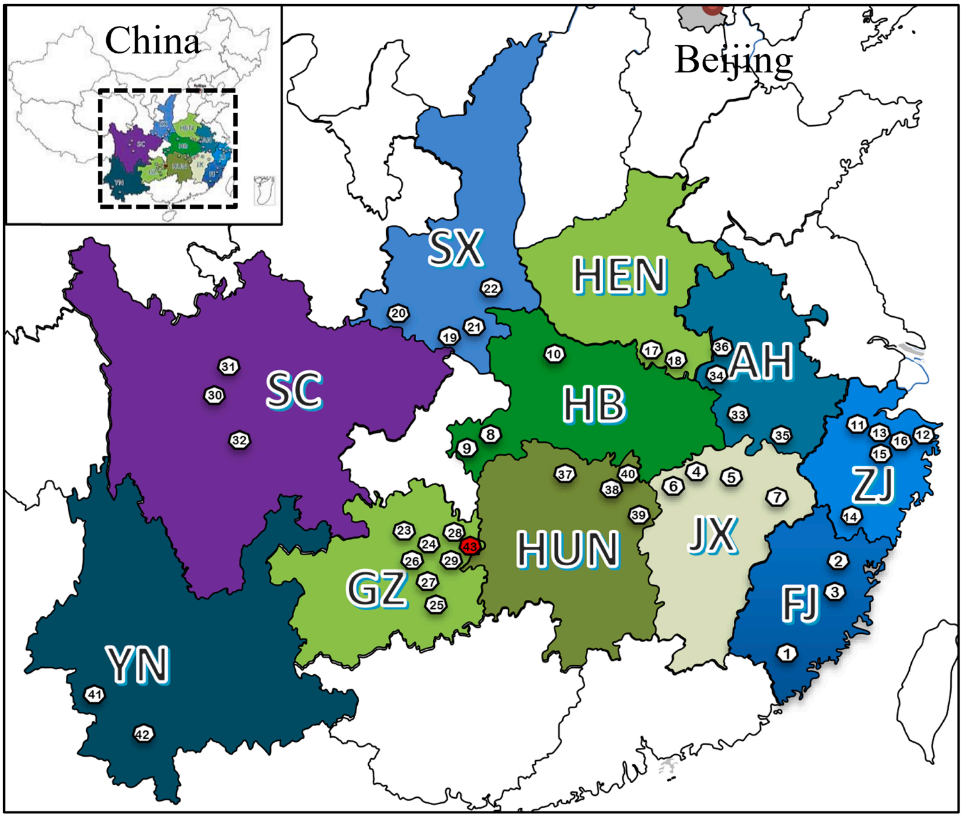

Fig. 1. The green tea sampling sites in different provinces in China (Fujian (FJ), Jiangxi (JX), Hubei (HB), Zhejiang (ZJ), Hunan (HUN), Shaanxi (SX), Guizhou (GZ), Sichuan (SC), Anhui (AH), Henan (HEN), and Yunnan (YN)).

Tekran Instruments, Canada) as per the United States Environmental Protection Agency (Method 1631E) (Du et al., 2021). Method blanks, certified reference materials (CRMs; GBW10020 - citrus leaf, 150 ng/g; GBW10048 - celery, 14.6 μg/kg), and duplicates were used for quality control. The mean recoveries of the CRMs were 97 ± 9% (N = 6) and 93 ± 7% (N = 6) for GBW10020 and GBW10048, respectively. The relative percentages of difference were less than 10% for duplicate samples.

The Hg leaching ratio was obtained as the Hg retained ratio in tea brew (η_1) by dividing the Hg leached in the tea brew with the total mercury in the tea (Karimi et al., 2008; Zhang et al., 2013). Due to the low Hg content in green tea and consequently, the amount of Hg entering the tea brew is relatively low, to ensure accuracy in this study, the Hg leaching ratio from tea is obtained by calculating the average of the Hg retained ratio in tea brew ($\eta_1 = (c_i \times v_i) / (c_t \times m_t)$), and the Hg lost ratio from tea leave ($\eta_2 = \{[(c_t \times m_t) - (c_r \times m_r)]\} / (c_t \times m_t)$). Ideally, the loss ratio of Hg from tea leaves (η_2) and the Hg retained ratio in tea brew (η_1) should be equal. Therefore, the Hg leaching ratio was calculated according to the equation below:

$$\eta = (\eta_1 + \eta_2)/2 = \{[(c_t \times m_t) - (c_r \times m_r)] + (c_i \times v_i)\}/(2 \times c_t \times m_t)$$

η, η_1, η_2 are the Hg leaching ratio from tea, the Hg retained ratio in tea brew, and the Hg lost ratio from tea leaves, respectively; c_r, c_i, and c_t are Hg concentrations in tea residues, tea infusions, and tea, respectively; m_r and m_t are the mass of tea residues and tea; v_i is the volume of tea infusions.

2.4. Dietary exposure and risk estimates

The dietary exposure and risk estimate method has been used in previous studies (Wang et al., 2020; Xu et al., 2020). The probable daily intake (PDI) for the general adult population was calculated using the following equation:

$$PDI = C_{THg} \times IR \times \gamma \times \varphi / bw$$

Where PDI is mentioned in micrograms per kilogram of body weight per day (μg/kg bw/day); bw = 60 kg; C_{THg} is the Hg concentration of green tea (μg/kg); IR is intake rate (g/d), the average tea consumption amount in China was considered to be about 0.3 kg/ year (Guan and Yang, 2014); γ is the Hg leaching ratio of green tea; φ is the proportion of green tea accounting for total tea consumption, in this study, the ratio was 53.25% according to the Analysis Report on the Development of Chinese Tea Industry (2019).

The total Hg exposure due to green tea consumption was estimated by using the hazard quotient (HQ) (Xu et al., 2020). The HQ value was calculated by comparing the PDI with the recommended probable tolerable weekly intake (PTWI) according to the equation below:

$$HQ_{THg} = 7 \times PDI/PTWI$$

Where the PTWI value is 4 μg/kg bw/week, it is recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA, 2010).

3. Results and discussion

3.1. Estimation of mercury content in green tea

According to Fig. 2a, more than 90% of green tea samples were found to contain < 10 μg/kg of Hg on dry weight basis. The average Hg content in green tea was 6.3 ± 6.4 μg/kg (in the range of 1.8–102.9 μg/kg dry weight), which was much lower than the maximum permissible limit (100 μg/kg) recommended the European Commission Regulation for certain contaminants in food items (2008) (Gasser et al., 2009), the European Pharmacopeia Monographs of herbal drugs (Martín-Domingo et al., 2017) and by the Ministry of Agriculture for heavy metals limited

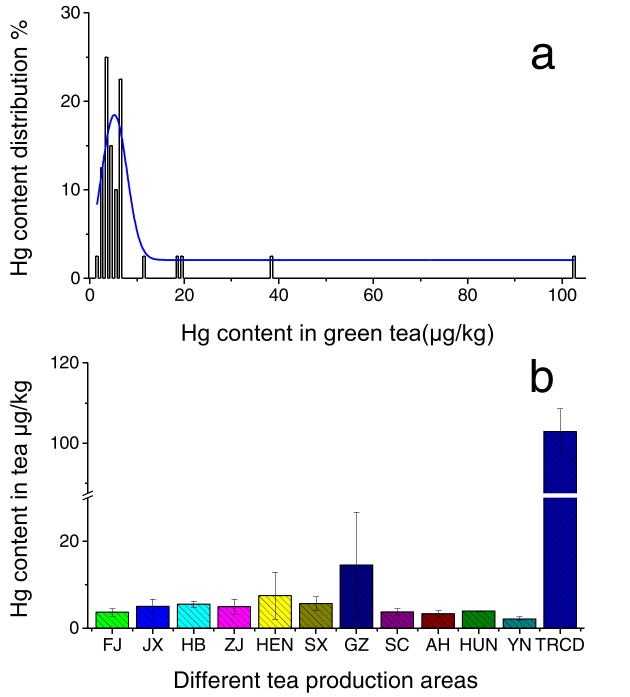

Fig. 2. (a)Frequency of mercury concentration in green tea in the representative samples and (b)average Hg concentration for different sampling areas: Fujian (FJ), Jiangxi (JX), Hubei (HB), Zhejiang (ZJ), Hunan (HUN), Shaanxi (SX), Guizhou (GZ), Sichuan (SC), Anhui (AH), Henan (HEN), Yunnan (YN)) and Tongrenchadian (TRCD).

standards (300 μg/kg) in tea in China (NY659, 2003). Only one green tea sample (TRCD) from a tea garden located in WMMA in Guizhou (Fig. 1, site 43) slightly exceeded the maximum Hg limit (102.9 ± 5.7 μg/kg) of European regulations for foodstuffs and herbal drugs. Except for the control sample (TRCD, sampling site 43), the highest Hg content was detected in green tea from Guizhou (GZ) (14.6 ± 12.2 μg/kg), which was about seven times higher than that from Yunnan Province (YN, 2.1 ± 0.5 μg/kg) and about twice than that from Henan Province (HN, 7.5 ± 5.4 μg/kg). The tea-producing areas in the sampling site of Guizhou (central and northern Guizhou) and Henan are close to Hg mining areas or located in the areas with severe Hg pollution (Wu et al., 2020a; Xie et al., 2012), resulting in higher Hg content in tea leaves from these sites. Our results were consistent with other studies that have reported that Hg levels in green tea usually are far below the maximum permissible limit (100 μg/kg), ranging from a few to dozens μg/kg (Table 1) (Árvay et al., 2015; Brodziak-Dopierała et al., 2018; Liu, 2009; Prkić et al., 2018; Schulzki et al., 2017; Wang et al., 2008; Zhang et al., 2012).

The accumulation of Hg in leaves increases with the increase in leaf age (Assad et al., 2016; Laacouri et al., 2013). Particularly, in the control sample collected from site 43, the Hg content was as high as 102.9 ± 5.7 μg/kg. Therefore, it is important to ascertain whether tea leaves in areas with high Hg contamination already contain extremely high levels of Hg once leaves sprout, or they accumulate for a short period between sprouting and picking. The results of Hg accumulation in tea leaves from the control site (sampling site 43) are shown in Fig. 3.

Leaves have long been recognized as a sink for atmospheric Hg, and the contribution of soil to leaf Hg has been deemed marginal (Laacouri et al., 2013; Liu et al., 2021b). The total gaseous Hg concentrations in WMMA were found to be in the range of 24–23,842 ng/m^3 (Chang et al., 2021) and were far more than the background value (1–9 ng/m^3) (Wu et al., 2020b) in China. The dry and wet deposition fluxes of Hg in WMMA also reached extremely high levels (Aslam et al., 2022). This suggests that Hg may accumulate to high levels in tea leaves in a relatively short period.

Table 1
Mercury content (μg/kg) in green tea reported in earlier studies.

Country	Number of samples or sample sites	Range of mercury concentration (μg/kg)	Reference
China	129	1.8–102.9 Average 6.3 ± 6.4 μg/kg	This study
China	5	2.37–4.16	(Árvay et al., 2015)
Japan	8	1.19–4.45	
Nepal	1	2.26	
Canada	41	< 30	
China	4	10–20	(Schulzki et al., 2017)
Vietnam	1	20	
Guizhou, China	3	22–31	(Zhang et al., 2012)
China	15	6.7–19.3	(Wang et al., 2008)
Japan	5	2.2–9.3	
China	6	2.7–5.0	(Liu, 2009)
Croatia	5	1.05–2.80	(Prkić et al., 2018)
Ukraine	1	1.55	
Austria	1	2.33	
Russia	1	1.05	
Polish	–	2.96–27.22 Average 13.39	(Brodziak-Dopierała et al., 2018)

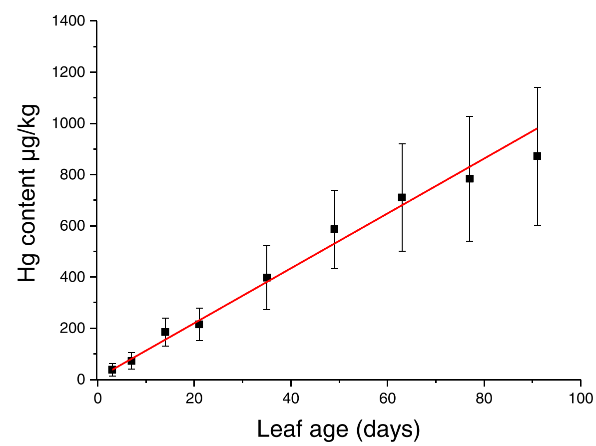

Fig. 3. The relation between mercury accumulation in tea leaves (from the control sampling site No. 43) and time.

As shown in Fig. 3, the concentrations of Hg in tea samples increased steadily in 90 days after sprouting, although its content was relatively lower in the first 10 days. The Hg accumulated in the leaf followed the equation: $y = 6.41 + 10.71x$, where y is the Hg content (μg/kg) in the tea leaf, and x is the leafage (days). This suggests that the presence of Hg in tea is mainly due to its accumulation during growth, and the earlier the tea leaves are picked, the lower the Hg content in tea. While, owing to the preference for particular tea brands, some people consume tea products of the same origin for a longer leaf growth time, posing a higher risk of Hg exposure.

4. Factors influencing mercury leaching during tea brewing

4.1. Mercury leaching in typical conditions

Hg leaching in tea brewing was carried out in single (one-time) and multiple brewing; these are also the main daily brewing ways practiced in China. Single (one-time) brewing was conducted only once, and the leaching ratio of Hg was examined during the natural cooling process (Fig. 4a). For multiple brewing, tea was brewed several times to examine the effect of different rounds of brewing on the Hg leaching ratio (Fig. 4b). For single brewing, the Hg leaching ratio increased sharply

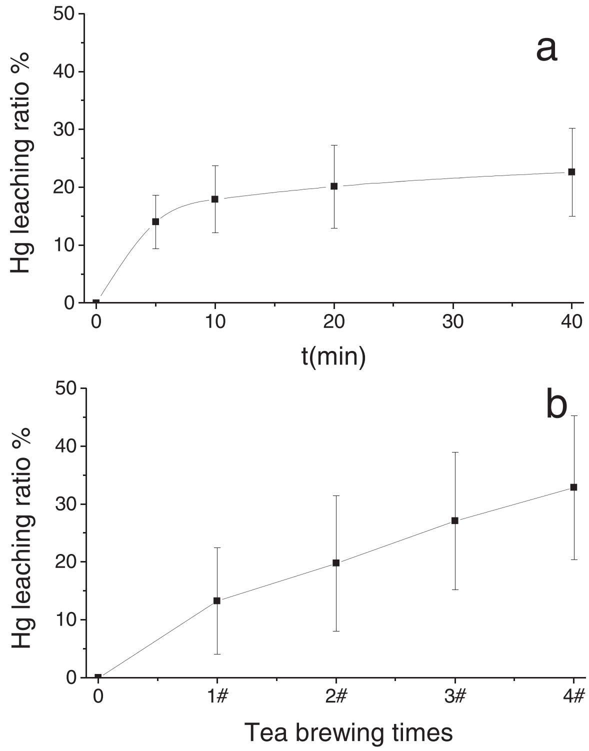

Fig. 4. The leaching ratio of mercury in green tea under typical conditions. (a) Single brewing and (b) multiple (four-times) brewing, each for 3 min At an initial water temperature of 97 °C, a solid-liquid ratio of 1:75, and room temperature (23 °C).

(14.00 ± 4.65%) in the first 10 min (Fig. 4a) and then slowly increased. The leaching ratio of a single brew reached 22.61 ± 7.58% in 40 min. In comparison, the leaching ratio of Hg during multiple brewing consistently increased with the number of brews (each brew lasted for 3 min). In the first brew, the leaching ratio reached 13.26 ± 9.23% and then increased at an average of about 6.52% for each brew. In four brews, the leaching ratio of Hg was up to 42.97% (average about 32.83 ± 12.37%). Thus, multiple brewing may increase the leaching ratio of Hg, thereby increasing the risk of Hg exposure if the daily consumption of tea is certain.

4.2. The factors affecting mercury leaching

To clarify the factors that affecting the Hg leaching from green tea, the solid/liquid ratio, temperature, and water quality (hardness) on the Hg leaching ratio were examined and the results are discussed below. The tea selected for testing was a sample of tea from the highest Hg content origin in each province including sampling sites 2, 6, 8, 12, 17, 20, 29, 30, 36, 37, 41 and 43 for a total of 12 samples.

4.3. The influence of water temperature

Fig. 5a shows that the water temperature could increase the Hg leaching ratio in green tea. As the brew temperature increased from 67 °C to 87 °C, the Hg leaching ratio increased slightly from 8.61% to 10.24% in 40 min of a single brew and from 14.00% to 22.61% after brewing four times. The Hg leaching ratio increased sharply when the temperature increased to 97 °C. Brewing temperature was found to be one of the key influencing factors on the leaching of green tea content (Jin et al., 2019; Sharpe et al., 2016). Jin et al. (Jin et al., 2019) suggested a significant difference in tea infusion content at different brewing temperatures. As the brewing temperature increases, substances including tea polyphenols, caffeine, and amino acids in tea infusions increase significantly (Chen et al., 2020). Hg may combine with these substances and leach in tea infusion.

4.4. The effect of solid/liquid ratio on mercury leaching ratio

Fig. 5b shows that the leaching ratio of Hg from green tea increased with the decrease in solid/liquid ratio to a certain extent regardless of the number of brews. The average leaching ratio increased from 18.27% to 25.69% in 40 min after one-time brewing and from 29.54% to 39.48% after brewing four times.

4.5. Influence of water quality on mercury leaching

The addition of calcium carbonate to the deionized water simulates the hardness of the water (Lung et al., 2008). Fig. 5c shows that the calcium carbonate in water has only little effect on the ratio of Hg leaching from green tea during the brewing process even with a high degree of hardness.

4.6. The risk of mercury exposure

A close relation was observed between Hg exposure risk (Fig. 6) via green tea consumption and Hg content in green tea from different producing areas. Except for the control sampling sites (sample site 43, TRCD), green tea from some sites in Guizhou may pose a higher Hg exposure risk, although, with HQ of only 6.11 ± 5.11%, its exposure risk was much lower than the critical value. The average HQ value of green tea consumption in China was only 1.82 ± 1.85% for one-time brewing for 40 min and 2.64 ± 2.68% for multiple (four-times) brewing. This result suggests that Hg exposure risk from green tea consumption is extremely low in most cases. Multiple brewing of tea leaves releases more Hg into the tea infusion, increasing Hg exposure risk. The HQ value of the green tea from sampling site 43 (TRCD) was 29.70 ± 1.66% after one-time brewing and 43.12 ± 2.41% after multiple (four-times) brewing. Thus, green tea, which has a low potential for Hg accumulation, may also pose a relatively high risk of Hg exposure in some cases. Although the HQ value is still less than 1 and its intake is still in the safe range, the risk of Hg exposure through green tea from the areas with high Hg pollution should not be ignored.

5. Conclusions

In this study, the Hg content in green tea from different production areas was determined and its leaching character during tea brewing was analyzed. Hg content in different green tea samples from China was found to be low, with an average content being 6.3 ± 6.4 μg/kg, which was significantly lower than the maximum permissible limit (100 μg/kg) specified by the Chinese food safety standards. This may be because green tea is normally produced from young leaves. Even in areas with extremely high Hg pollution, its content in green tea can still be controlled to very low levels by managing the leaf-picking period. The Hg leaching ratio after one-time brewing was 22.61 ± 7.58% in 40 min and was up to 42.97% (average of 32.83 ± 12.37%) after multiple (four-times) brewing. An increase in brewing temperature, frequency, and solid/liquid ratio was found to promote the leaching of Hg, while water hardness had only a little effect. Thus, the potential for Hg accumulation in green tea and Hg exposure to humans from green tea consumption is extremely low, although the risk may be relatively high when tea leaves are picked from high Hg pollution areas.

CRediT authorship contribution statement

Qingfeng Wang: Methodology, Investigation, Data curation, Formal analysis, Writing - original draft. **Dan Wang**: Investigation, Software, Visualization. **Zhonggen Li**: Conceptualization, Writing. review & editing. **Yuyu Wang**: Investigation. **Yan Yang**: Investigation. **Mengxun**

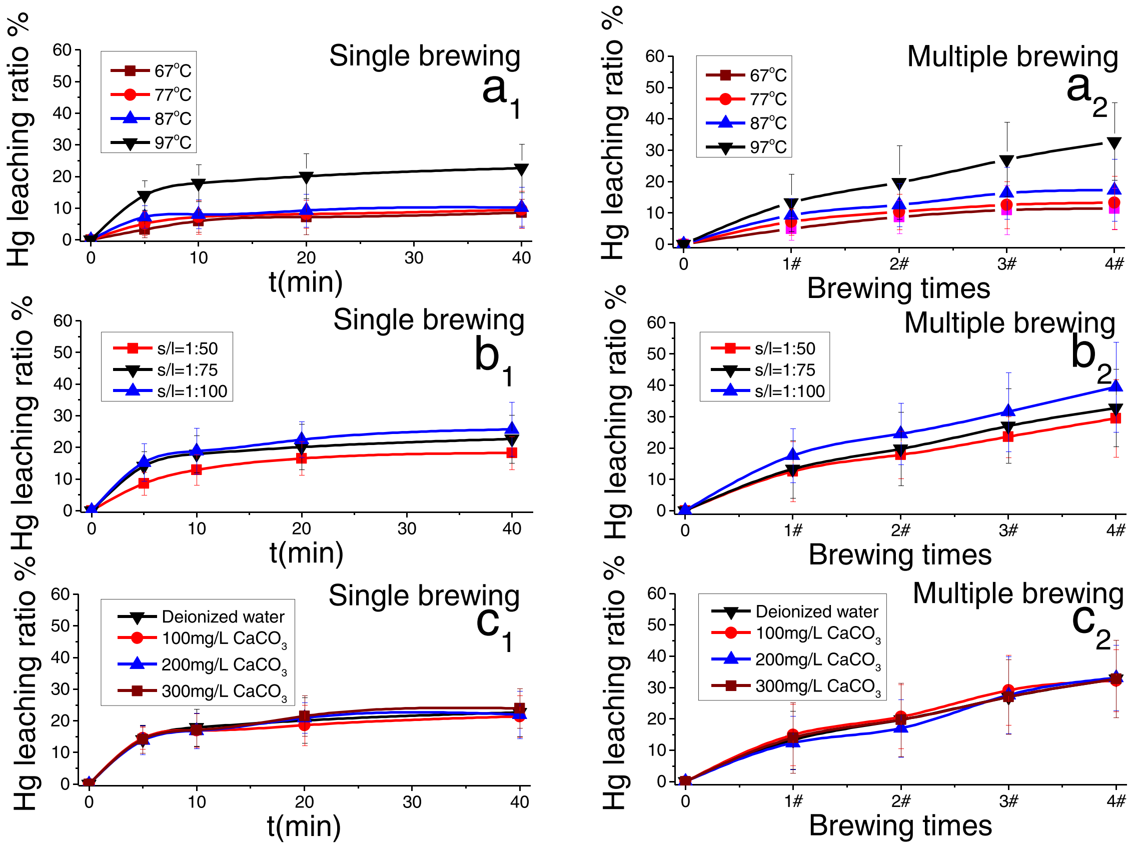

Fig. 5. Effects of brewing conditions on Hg leaching ratio, a. the effect of temperature (Single (one-time) brewing (a_1) and four-times brewing with each step lasting 3 min (a_2) at a solid-liquid ratio of 1:75 and room temperature (23 °C)); b. the effect of solid/liquid ratio (Single (one-time) brewing (b_1) and multiple (four-times) brewing, each for 3 min (b_2) at an initial temperature of 97 °C and room temperature (23 °C)); c. The effect of different concentrations of calcium carbonate (single (one-time) brewing (c_1) and multiple (four-times) brewing, each for 3 min, (c_2) at an initial temperature of 97 °C, a solid-liquid ratio of 1:75, and room temperature (23 °C)).

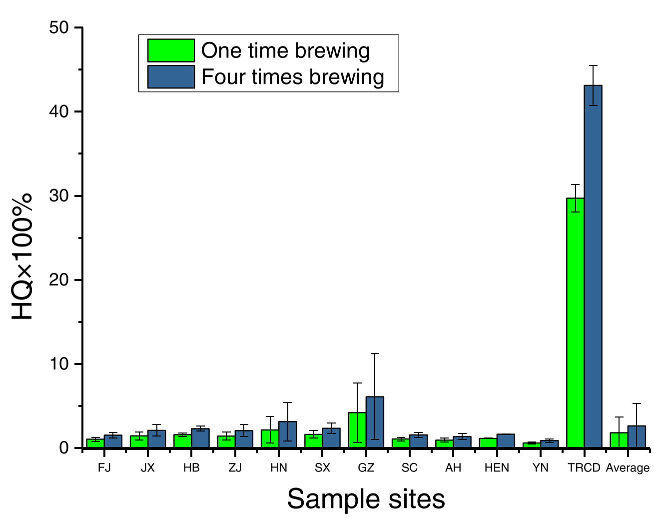

Fig. 6. Hazard quotient (HQ) values of the green tea intake from different sampling sites (Fujian (FJ), Jiangxi (JX), Hubei (HB), Zhejiang (ZJ), Hunan (HUN), Shaanxi (SX), Guizhou (GZ), Sichuan (SC), Anhui (AH), Henan (HEN), Yunnan (YN)) and Tongrenchadian (TRCD)).

Liu: Investigation. **Dadong Li:** Investigation. **Guangyi Sun**: Writing. review & editing. **Boping Zeng**: Resources, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2022.113279.

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