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Occurrence, spatial distribution, and risk assessment of perchlorate in tea from typical regions in China

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ABSTRACT

Perchlorate is a kind of persistent pollutant which occurs widely in the environment. The news of “high content of perchlorate detected in tea exported from China to Europe” has aroused public concerns on perchlorate in tea. However, limited data on its occurrence in tea and health risks for the tea consumers are available. To this end, this study explored the occurrence and spatial distribution of perchlorate based on 747 tea samples collected from the 13 major tea producing regions in China. Perchlorate was detected in 100% of tea samples. The average concentration of perchlorate was 163 μg/kg with the range from 1.2 μg/kg to 3132 μg/kg. From the perspective of spatial distribution, a remarkable difference was observed for perchlorate concentrations in tea samples between different regions ($p < 0.0001$), and the average concentration of perchlorate from the central China (409 μg/kg) was higher than that from the eastern (90.7 μg/kg) and western (140 μg/kg) regions. However, this study cannot obtain the difference of perchlorate concentrations between different tea categories. Furthermore, a human exposure assessment of perchlorate intake through tea consumption was performed by deterministic and probabilistic risk assessment. The average chronic daily intake (CDI) to perchlorate of Chinese tea consumers was 0.0183 μg/kg bw/day, however, CDI for high tea consumers (99% and 99.9%) was 0.1514–0.4675 μg/kg bw/day. The health risk assessment conducted with a hazard quotient showed that perchlorate exposure through tea consumption was under a safety threshold. Nevertheless, if other dietary exposure pathways were considered, health risks to perchlorate for high tea consumers would be paid attention to.

1. Introduction

Perchlorate (ClO_4^-) is a water soluble and highly stable persistent environmental pollutant. The formation of perchlorate can occur by spontaneous generation in the environment, however, the occurrence of perchlorate in the environment is mainly caused by human activities (Murray and Bolger, 2014). Perchlorate is often used as a strong oxidant in rocket propellants, pyrotechnic manufacturing, arms industry, blasting operations, and other fields (FAO/WHO, 2011). It can also be used as an additive in the production process of rubber products, dye coatings, lubricants, and other products (Vella et al., 2015). Because of its wide application and ubiquitous occurrence (Pleus and Corey, 2018), once perchlorate enters the environment, it will rapidly spread to both groundwater and surface water, leading to further expansion of pollution (Cao et al., 2019). Plants can absorb and enrich perchlorate from water and soil, and finally enter the human body through the food chain (Kumarathilaka et al., 2016). At present, perchlorate was detected in vegetables, fruits, dairy products, aquatic products, meats, and other food categories (Abt et al., 2018; Chen et al., 2021; Chen et al., 2022; Chen et al., 2023; EFSA, 2017; Kim et al., 2014; Lee et al., 2012; Li et al., 2022; Liao et al., 2020; Wang et al., 2021), which has aroused widespread concerns. Other exposure pathways of perchlorate also include

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ingestion and inhalation of indoor dust, as well as atmospheric dust (Jiang et al., 2021; Wan et al., 2015).

The effect of thyroid function is considered to be the most important concern of perchlorate on human health (Leung et al., 2010). Since the size and charge of perchlorate are very close to iodine ions, it can compete with iodine to enter the thyroid gland of the human body, and interfere with the absorption of iodide by the sodium iodide symporter. However, inadequate iodine uptake can affect the synthesis and release of thyroid hormones T3 and T4, leading to the degradation of thyroid function (Abt et al., 2018). To protect human health, the National Environmental Protection Agency issued a daily reference dose of 0.7 μg/kg bw/day for perchlorate in 2005 according to the recommendations of the National Academy of Sciences (NAS, 2005).

Tea is the second largest non-alcoholic consumption beverage in addition to water, mainly possessing flavor, aroma, and potential health benefits (Bag et al., 2022; Wang et al., 2023). According to the tea production process, it is generally divided into green tea, black tea, oolong tea, white tea, black tea, and yellow tea. Perchlorate has a high bioaccumulation rate in leafy plants (Liao et al., 2020). The intake of perchlorate in tea mainly depends on variety, plant growth time, weather conditions, and perchlorate concentration (Liao et al., 2022). In recent years, the news of "high content of perchlorate detected in tea exported from China to Europe" has aroused public concerns on perchlorate in tea. The European Union (EU) has set the reference residue limit of perchlorate in dried tea as 0.75 mg/kg (EC, 2015), and adds perchlorate into the annex of EU regulations on food contaminant limits from July 1, 2020. Since perchlorate is a new kind of pollutant, China has not established a food safety limit standard for perchlorate in tea.

So far, few studies have been conducted on the contamination level of perchlorate in tea samples (EFSA, 2017; Liao et al., 2022; Yao et al., 2022). Limited data are available on the exposure assessment to perchlorate for tea consumers (Liao et al., 2022; Yao et al., 2022). Given a negative impact of perchlorate for the tea industry and its widespread occurrence in the environment, a more extensive understanding on the occurrence in tea and health risks to tea consumers is required. Therefore, this study further expanded the number of tea samples ($n = 747$) collected from 13 major tea producing regions in China to obtain more refined baseline contamination level of perchlorate. Additionally, the spatial distribution trend of perchlorate pollution was explored, and the health risks of tea consumers were assessed through deterministic and probabilistic risk assessment.

2. Chemicals and methods

2.1. Chemicals and reagents

Perchlorate standard solution ($Cl^{16}O_4^-$, 1003 ± 4 mg/L) was purchased from Inorganic Ventures (NJ, USA). $Cl^{18}O_4^-$ solution (100 mg/L) obtained from Cambridge isotope laboratory (MA, USA) was used as the internal standard. Reagents (LC-grade) used in this experiment, such as acetonitrile, methanol, and acetic acid, were obtained from Fisher Scientific (TX, USA). Ultra-pure water was prepared using the Milli-Q system (Millipore, MA, USA).

2.2. Sample collection

During 2017–2019, a total of 747 tea samples were randomly collected from the markets of 13 major tea producing regions, including Fujian ($n = 349$), Anhui ($n = 61$), Hunan ($n = 30$), Zhejiang ($n = 89$), Yunnan ($n = 59$), Sichuan ($n = 19$), Shaanxi ($n = 14$), Jiangxi ($n = 21$), Jiangsu ($n = 15$), Hubei ($n = 25$), Henan ($n = 15$), Guangxi ($n = 25$), and Guangdong ($n = 25$). According to the known tea categories, these tea samples were divided into the following six types: green tea ($n = 189$), dark tea ($n = 43$), black tea ($n = 90$), oolong tea ($n = 234$), white tea ($n = 56$), and yellow tea ($n = 25$). Additionally, to further evaluate the contamination of perchlorate among different larger regions in China, this study divided Fujian, Zhejiang, Jiangsu, and Guangdong into the Eastern China, Anhui, Hunan, Jiangxi, Hubei, and Henan as the Central China, and Yunnan, Sichuan, Guangxi, and Shaanxi as the Western China. Each tea sample (approximately 300 g) was ground into fine powder and stored in a refrigerator at −20 °C before analysis.

2.3. Sample preparation

In this study, the sample preparation of perchlorate was based on the analytical method we previously reported (Li et al., 2020), and slightly modified. Briefly, exactly 1.0 g of tea sample was weighed into a 15 mL polypropylene tube and 20 μL of the isotope internal standard $Cl^{18}O_4^-$ solution (1.0 mg/L) was spiked. After the sample was homogenized for 30 min, 6 mL of water was added to soak for 30 min, and then 4 mL of acetonitrile was added, and ultrasonic extraction was performed. After centrifugation, the supernatant was treated for cold-induced phase separation at - 30 °C for 45 min. Then, the upper phase was diluted with the same volume of water and centrifuged at 13,000 rpm for analysis.

2.4. Instrumental analysis

Liquid chromatography - high resolution mass spectrometer (Thermo, Bremen, Germany) was used for the determination of perchlorate. As for chromatographic analysis, perchlorate and internal standard $Cl^{18}O_4^-$ were separated on a Poroshell 120 pentafluorophenyl column (50 × 2.1 mm, 1.9 μm) under a gradient elution program. The temperature of column oven was set at 40 °C. The mobile phase was composed of methanol (A) and 1% of acetic acid aqueous system with the following program: 0–0.8 min, 10%A; 0.8–2.2 min, 10%–100%A; 2.2–2.5 min, 100%A; 2.5–4.0 min, 100%–10%A. The flow rate and injection volume were set as 0.6 mL/min and 10 μL, respectively. As for detection analysis, instrument was tuned at a heated negative electric spray ionization with the following parameters: spray voltage, capillary temperature, and heated temperature were set as 2.8 kV, 320 °C, and 450 °C, respectively. Meanwhile, the target single ion monitoring mode was used to acquire the raw data with 70,000 FWHM of resolution. The automatic gain control was set as 2e5, the maximum injection time was set as 100 ms, and the separation width was set as 5 Da. In the instrumental analysis, the quantitative ion of perchlorate was selected as m/z 98.9485, and the qualitative ion was selected as m/z 100.9456. The quantitative ion of internal standard $Cl^{18}O_4^-$ was for m/z 106.9680.

2.5. Quality control and quality assurance

To avoid system contamination and ensure the method selectivity, a solvent blank and a blank tea sample were used to ensure the feasibility and reliability of the method. The blank green tea sample was obtained from the Inter-laboratory Comparison Study on Perchlorate in Tea of the EU-China-Safe project (EU H2020 No. 727864). Of note, the microporous filter membrane used for sample solution filtration in this study would produce system residues of perchlorate. Thus, the sample analytical solution in this experiment was not filtered by microporous membrane, but centrifuged at 13,000 r/min for 5 min to avoid the background interference of perchlorate and eliminate the occurrence of false positive results. In real samples analysis, a system solvent blank was added along with every 20 samples to ensure the method reliability. A series of perchlorate standard solutions (0.1, 0.2, 0.5, 1, 2, 5, 10, 20, and 50 μg/L) were prepared to determine the concentration of perchlorate in tea. The correlation coefficient for the standard curve was greater than 0.999. The limit of detection and quantification were 0.2 and 0.6 μg/kg, respectively. Additionally, blank tea samples were selected for spiked recovery test as the method validation. The selected spiked levels were set at 1, 10, and 100 μg/kg. Each spiking level was measured in six times on a day for three consecutive days. The results showed that the recovery rate of perchlorate in the spiked blank tea was 82.7%–96.4%, while the intra-day precision and the intra-day precision was determined to be 0.8%–7.6% and 2.7%–11.8%, respectively.

2.6. Deterministic and probabilistic risk assessment

Tea has been reported to be one of the food categories with the highest residue level of perchlorate (EFSA, 2017), and has a unique tea drinking population in China. Therefore, perchlorate intake may have some health harm for tea consumers. In this study, the health risk of perchlorate was mainly evaluated for the main tea drinkers (Chinese adults) based on the China National Nutrient and Health Survey. The chronic daily intake (CDI) of perchlorate via the tea intake pathway can be calculated as Equation (1):

$$CDI = \frac{C \times ID \times TR}{bw} \tag{1}$$

where C is the perchlorate concentration in tea (μg/kg), ID is the consumption intake data of tea (g/day), TR is the transfer rate of perchlorate from dry tea to tea infusion, and bw is the body weight (kg). As for deterministic risk assessment, point estimates were used based on the average concentration level of perchlorate and tea consumption data. Tea consumption raw data and body weight of tea consumers were obtained from China National Nutrition and Health Survey by a 24-h recall in 2002 (Cao et al., 2018). In the tea consumption data, the daily average consumption of dry tea was 7.91 g, ranging from 0.3 g to 61.9 g. The standard body weight of Chinese adults (63 kg) was used. Additionally, the transfer rate of perchlorate from dry tea to tea infusion was obtained from a recent study, with a range of 0.589–0.892 (Liao et al., 2022).

According to the probabilistic risk assessment guidance method (EFSA, 2012), probabilistic assessments were performed by @Risk® 8.0 software based on Monte Carlo simulations, including distribution fittings, calculations, and random samplings. Detailed descriptions can be found in the Supplementary Materials and our previous study (Ren et al., 2023).

2.7. Statistical analysis

SPSS 18.0 and GraphPad Prism 7.04 were used for data analysis. Considering that the data obtained in this study is not normally distributed, the Kruskal–Wallis test was performed for comparative analysis of perchlorate concentrations in different regions and different types of tea, while Tukey's multiple comparisons test was used for difference analysis between the two. The value of $p < 0.05$ was considered statistically significant.

3. Results and discussion

3.1. Concentration and detection frequency of perchlorate

A total of 747 tea samples collected from 13 major tea producing regions in China were determined to obtain perchlorate concentrations, and detailed statistical analytical results were presented in Table 1. In this study, the detection frequency of perchlorate was 100%, which was basically consistent with previous studied results, such as, that of Chang study (100%, $n = 11$) (Chang et al., 2020), Yao study (99.3%, $n = 286$) (Yao et al., 2022), Zhao study (95.4%, $n = 240$) (Zhao et al., 2018), and Liao study (94.8%, $n = 288$) (Liao et al., 2022), indicating the widespread occurrence of perchlorate in the environment. The average concentration of perchlorate in monitored tea samples was 163 μg/kg (median: 77.0 μg/kg) with the concentration range from 1.2 μg/kg to 3132 μg/kg. For comparisons, Table S2 lists the perchlorate concentrations in tea from other studies. In a study of 288 tea samples collected from 16 regions of China (Liao et al., 2022), the average concentration of perchlorate was 295 μg/kg (range: non-detected (ND) −1274 μg/kg), while that was 350 μg/kg (range: 71.5–1603 μg/kg) in Chang study (Chang et al., 2020). Compared with these two studies, the average concentration of perchlorate in this study was relatively low. However, the average result in this study was equivalent to that of Fujian region (152 μg/kg) and Zhejiang region (187 μg/kg) from China (Yao et al., 2022; Zhao et al., 2018). In addition, a monitoring study in tea and herbs from the EU (EFSA, 2017) also showed a relatively high average perchlorate concentration (324 μg/kg, $n = 1193$). In our previous study (Chen et al., 2023), we compiled and summarized the residual levels of perchlorate in other dietary categories. As a result, it was found that the concentration level of perchlorate in tea was significantly higher than that in other food categories. Furthermore, the perchlorate residue in a 3.88% (29 of 747) tea samples in this study exceeded the maximum residual level of the EU (750 μg/kg), which was higher than that (2.08%) of the previous study (Liao et al., 2022).

3.2. Spatial distribution of perchlorate contamination

Considering that perchlorate is a ubiquitous environmental contaminant, the average concentration level of perchlorate in tea reported by various regions is still different, which is mainly related to the degree of perchlorate contamination in tea collected regions. To this end, perchlorate concentrations in tea collected from 13 major tea producing regions of China were analyzed and presented in Fig. 1A. The highest average concentration of perchlorate was observed in Hunan region (783 μg/kg), which was also consistent with that in a Liao study (Liao et al., 2022). However, the highest concentration of perchlorate in an individual tea sample collected in Hunan region reached 3132 μg/kg in this study, which was higher than that in Taiwan region (1603 μg/kg)

Table 1
Statistical analysis of detected perchlorate results from 13 regions (μg/kg).

Regions	N.	Min	25% Percentile	Median	75% Percentile	Max	Mean	Std. Deviation
Fujian	349	6.5	31.4	53.8	97.1	1007	79.0	91.3
Anhui	61	5.7	91.8	150	313	1345	263	290
Hunan	30	154	305	522	924	3132	783	689
Zhejiang	89	2.2	40.7	86.7	168	653	118	111
Yunnan	59	1.2	12.1	30.6	49.8	1361	73.1	218
Sichuan	19	80.5	121	144	232	466	184	101
Shaanxi	14	47.5	178	241	964	1155	462	402
Jiangxi	21	25.7	64.6	135	439	1364	313	357
Jiangsu	15	36.6	99.5	148	253	713	207	188
Hubei	25	122	249	401	573	1215	449	273
Henan	15	73.7	216	287	454	639	320	176
Guangxi	25	9.7	13.2	52.3	126	408	85.1	93.7
Guangdong	25	16.0	35.6	77.0	120	199	84.7	50.5
Total	747	1.2	35.5	77.0	172	3132	163	267

Note: N., the number of tea; Min, minimum; Max, maximum; Std. Deviation, standard deviation.

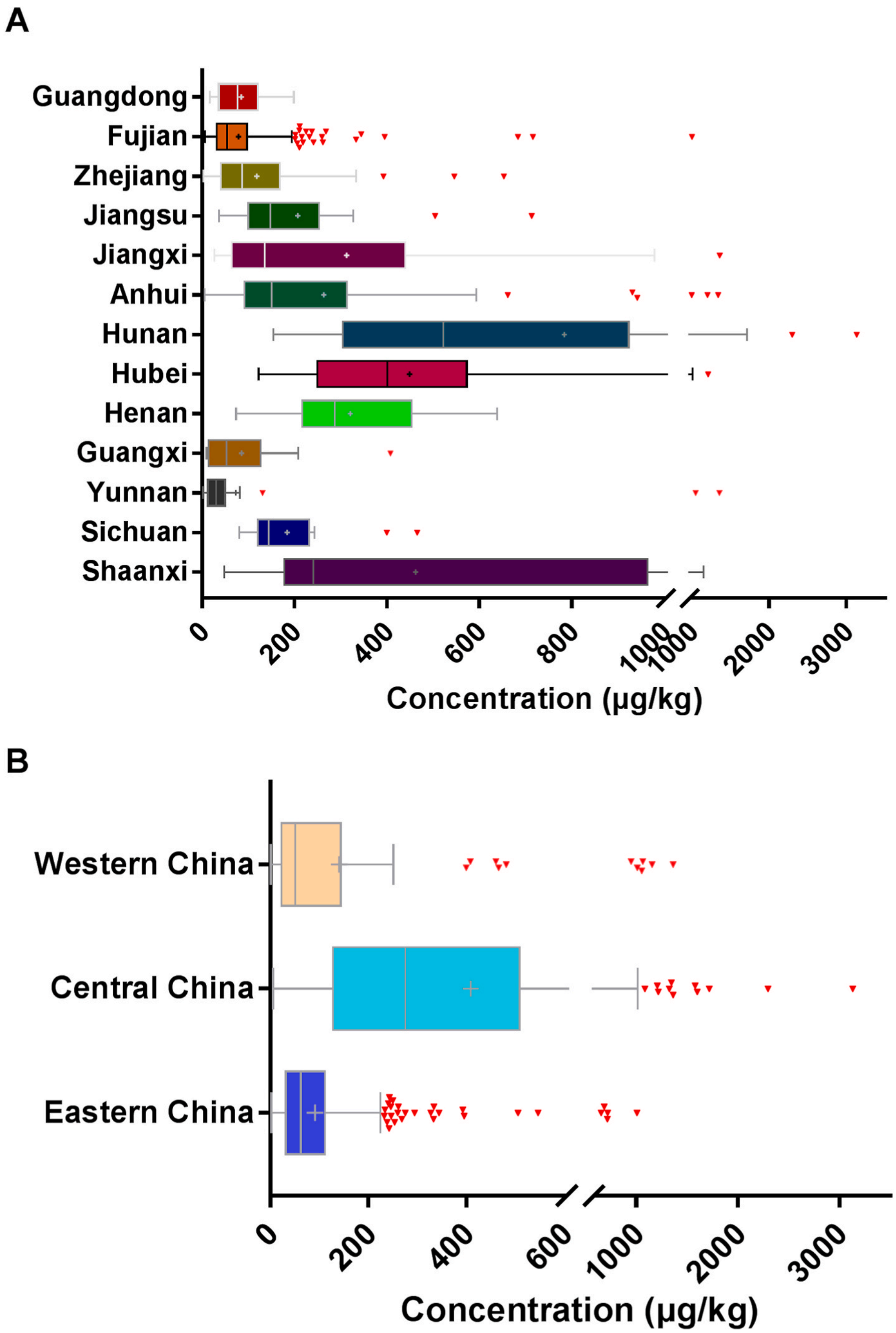

Fig. 1. Perchlorate concentrations in tea collected from (A) 13 major tea producing regions and (B) three main tea producing regions. Note: Concentrations were presented by box and whisker plot. The lower and upper bands of the box plot represent 25th and 75th of residue data, respectively, and the band inside the box indicates the median value. Box plot whiskers extend to 1.5 times the interquartile range. The red points indicate outliers; "+" indicates the mean value of concentration.

and Hunan region (1274 μg/kg) from other studies (Chang, et al., 2020; Liao et al., 2022). In this study, the average concentrations of perchlorate in Yunnan, Fujian, Guangdong, and Guangxi regions were relatively low with the values of 73.1, 79.0, 84.7, and 85.1 μg/kg, respectively. This studied result was also consistent with the previous reports that the perchlorate levels of tea samples in Zhejiang and Fujian regions of China were relatively low (Yao et al., 2022; Zhao et al., 2018). To further compare the difference of perchlorate concentrations in tea from different regions in this study, the Kruskal–Wallis test was performed from perchlorate residues between 13 regions with a significant difference ($p < 0.0001$), and detailed Turkey's multiple comparison results were presented in Table S3.

In this study, we divided the 13 monitoring regions into three main tea producing regions, namely eastern, central, and western China. As shown in Fig. 1B, The highest average concentration of perchlorate was observed in the central China (409 μg/kg, from 5.7 to 3132 μg/kg), followed by the western China (140 μg/kg, from 1.2 to 1361 μg/kg) and the lowest in the eastern China (90.7 μg/kg, from 2.2 to 1007 μg/kg), indicating that there is a remarkable difference of perchlorate concentrations among three main regions ($p < 0.0001$). However, no significant difference was observed from that of eastern and western China ($p > 0.05$). Tea plants can absorb perchlorate from the soil, irrigation water, and fertilizers, and inevitably some of which will be transported to the tea (Liao et al., 2022). Thus, perchlorate, as a ubiquitous environmental contaminant, may reflect the situation of perchlorate contamination in the local environment to a certain extent through the residual monitoring of perchlorate in tea. It can be seen from Fig. 2 and S1 that the mean and median perchlorate concentrations in tea in Chinese border and coastal regions, such as Yunnan, Guangxi, Guangdong, Fujian, Zhejiang, and Jiangsu regions, were relatively low compared with that in inland or central regions. Among them, the high content of perchlorate in central China is closely related to the surrounding environment, such as irrigation water, dust, and soil. Previous studies have also found that perchlorate is ubiquitous in indoor, outdoor, and soil in different regions of China (Gan et al., 2014, 2015; Li et al., 2018), and a relatively higher concentration of perchlorate was observed from fireworks manufacturing areas in China (Zhang et al., 2015). So, there is a certain correlation between the main production areas of fireworks and firecrackers in Hunan and Jiangxi in China and the exposure to perchlorate in these areas (Li et al., 2014).

3.3. *Concentrations of perchlorate from different tea categories*

Previous literature have reported that different types of tea can affect the final residual concentration of perchlorate due to the different time of picking fresh tea (Liao et al., 2022; Liu et al., 2019). Especially, dark tea may produce higher content due to the longer accumulation time of perchlorate. Table S4 shows the concentration distribution of perchlorate from the six different tea categories in this study. The average perchlorate concentrations for six tea categories were observed as the following trend: yellow tea (626 μg/kg) > green tea (224 μg/kg) > dark tea (172 μg/kg) > black tea (168 μg/kg) > white tea (105 μg/kg) > oolong tea (66.8 μg/kg). However, this trend was different from previous studies (Chang et al., 2020). The reason for this difference was related to the different perchlorate contamination in the regions where various types of tea were located, indirectly confirming that the contamination situation in each region was the leading factor of perchlorate contamination in tea. To this end, this study further compared and analyzed various types of tea (oolong tea, white tea, black tea, and green tea) collected from the same region (Fujian). Table S5 shows that the average concentration level of perchlorate in oolong tea was relatively low, and a significant difference ($p < 0.05$) between the content of perchlorate in oolong tea and other three types of tea was observed, which was different from that of the previous study also collected in Fujian region, which reported that oolong tea had a higher perchlorate residue (Yao et al., 2022). The reason for the difference in perchlorate content among different types of tea in Fujian may be closely related to the regions where tea is harvested. For example, there are two main source areas of oolong tea in Fujian, including Quanzhou and Mount Wuyi. In this study, Oolong tea in Fujian was mainly collected in Mount Wuyi area, which is related to the low level of perchlorate contaminant in the local area. In addition, the differential changes in perchlorate among different types of tea are also somewhat related to the tea harvesting season, which may not have been considered in this study and further attention needs to be paid in future study. Based on these studied results, there are still many uncertain factors to evaluate the contamination degree of perchlorate for six tea categories by simply relying on random sampling.

3.4. *Health risk assessment*

Here, the deterministic risk assessment of perchlorate exposure was first evaluated for the average tea consumption populations using the point estimates. The results showed that CDI on the average was 0.0183 μg/kg bw/day. Compared with RfD value of perchlorate, the HQ value was 2.61%. Understanding the exposure risk of perchlorate in high tea consumers, the estimated dietary intake to perchlorate via tea consumption was further evaluated using the probabilistic approach. Based on the results of Monte Carlo Simulations modelling 1, the CDIs of perchlorate for tea consumers at the 50th, 95th, 99th, and 99.9th percentile were 0.0047, 0.0578, 0.1514, and 0.4033 μg/kg bw/day, respectively, while for Monte Carlo Simulations modelling 2, these values were 0.0048, 0.0596, 0.1599, and 0.4675 μg/kg bw/day, respectively (Fig. 3 and Table S6). Compared with a RfD value of perchlorate (0.7 μg/kg bw/day), HQ values were less than 1.0 at all percentiles for all samples. Of note, HQ values at 99th and 99.9th percentiles for two modelling were 21.63–22.84% and 57.62–66.79%, respectively. These results showed minimal risks of exposure to

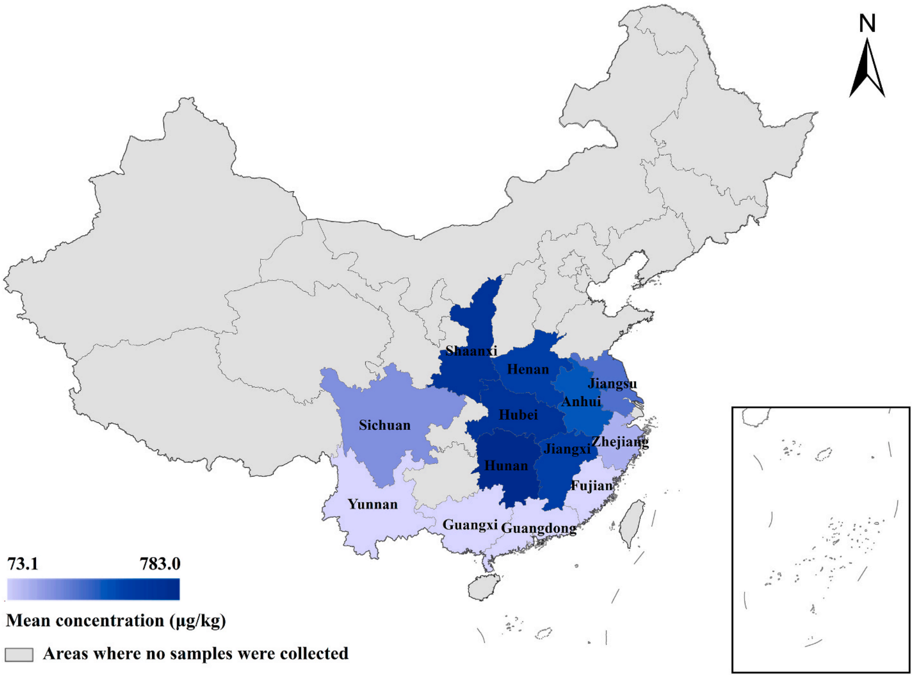

Fig. 2. Distribution of perchlorate (mean concentration) in tea collected from 13 major tea producing regions.

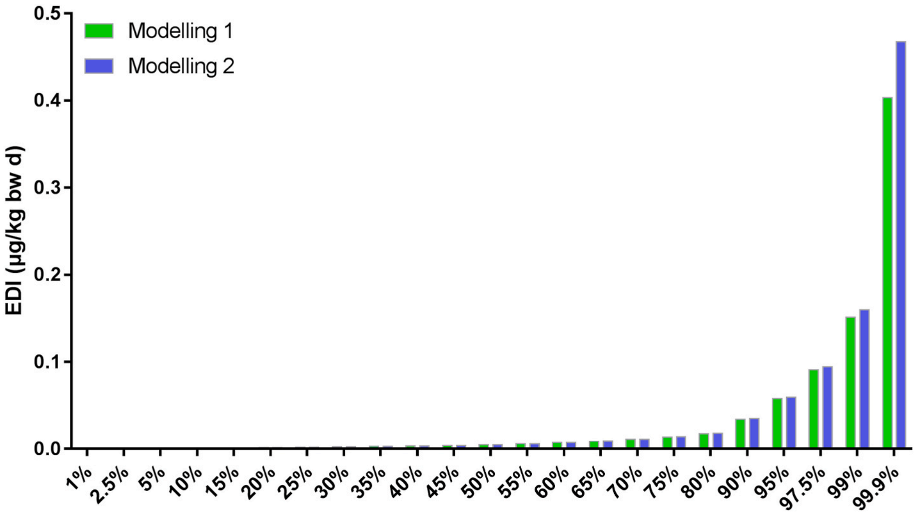

Fig. 3. CDIs of perchlorate for tea consumers from different percentiles based on the results of Monte Carlo Simulations.

perchlorate through tea consumption.

Comparison of perchlorate CDI (ng/kg bw/day) from tea consumption between this study and other studies was listed in Table S7. The estimated average CDI value of perchlorate in this study was two times lower than that in the study of Liao et al., but no significant difference was observed for the CDI value from the high tea consumption population (Liao et al., 2022), which may be related to the fact that the contaminant level of perchlorate in tea was a positive-skewed

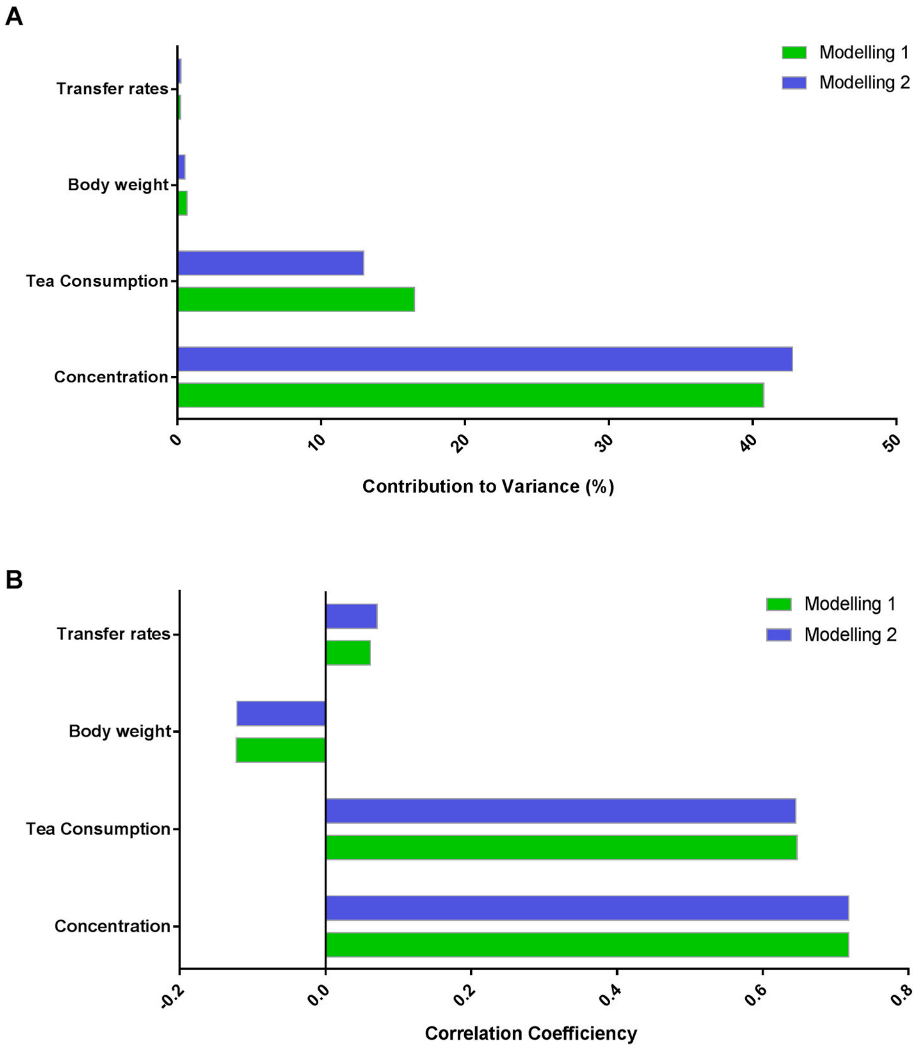

Fig. 4. Sensitivity analysis of the CDIs of perchlorate for the influencing variables using contribution to variance (A) and correlation coefficient (B).

distribution. The same phenomenon was also observed in Yao et al. study (Yao et al., 2022). As for the comparison of CDI values of perchlorate in tea and other food categories, we can refer to the literature data previously reported, which presented the detailed CDIs in other food categories from different countries and regions (Chen et al., 2023). On the whole, the contribution of tea to perchlorate in total dietary samples is relatively low.

This assessment shows that there is no need to carry out risk management on tea consumers due to minimal risks of exposure to perchlorate through tea intake. However, if other dietary exposure pathways are considered, some actions need to be taken to reduce the future exposure to perchlorate through tea intake for high tea consumers.

3.5. *Sensitivity analysis of the CDIs of perchlorate*

Sensitivity analysis was performed to identify the most important influencing variable that can determine the greatest impact on the estimated health risk by quantifying the contribution rate and correlation coefficient caused by each parameter in the model. Fig. 4A shows the results of sensitivity analysis to assess the chronic health risks of tea consumers exposed to perchlorate. Under these two modelling methods, the variance contribution for perchlorate concentration and tea consumption was ranged from 40.8% to 42.8% and 13.0%–16.5%, respectively, indicating that perchlorate residue and tea consumption were the main contributors. In addition, body weight and transfer rate had a negligible impact on the estimated risk of perchlorate exposure ($<1\%$). Therefore, reducing the concentration of perchlorate in tea can be conducive to reduce the chronic health risk for Chinese tea consumers. Furthermore, spearman correlation coefficient was also used to analyze the sensitivity of the distribution of various input parameters generated in risk assessment. Fig. 4B shows the correlation between input parameter distribution and the HQ. Among the two models, HQ had the strongest positive correlation with the perchlorate concentration ($r = 0.72$) and tea consumption ($r = 0.65$) in the risk assessment. Moreover, the transfer rate of perchlorate from dry tea to tea infusion was also positively correlated with HQ, but the correlation was weak ($r = 0.06$–0.07). On the contrary, the body weight of adults was inversely and weakly correlated with HQ ($r = -0.12$). These results further indicated that the individual's body weight and transfer rate had no direct impact on perchlorate exposure, so it would not affect the health risk.

3.6. *Strengths and limitations of this study*

The strengths of this study included that a much larger number of tea samples were collected from 13 major tea producing regions in China. This study first presented the spatial distribution of perchlorate contamination in 13 major tea producing regions in China, which can be referred to as the regional change of perchlorate contamination in these regions to a certain extent. In addition, compared with traditional point estimate methods based on uniform parameters, probabilistic risk assessment based on Monte Carlo simulation provided more refined estimates considering variable probability distribution, such as different tea consumption populations and consumption data. However, some limitations were presented in this study. First of all, the risk assessment of perchlorate in this study is only based on an individual tea intake, without considering the intake source of other food categories. It is necessary to combine exposure assessment with other food categories. Secondly, the tea consumption data used in the evaluation method of this study is from the China National Nutrition and Health Survey by a 24-h recall in 2002, so uncertainty may be introduced into the exposure assessment. Thirdly, the perchlorate concentration levels in tea s vary greatly with time and space. The different harvesting time of tea may affect the survey results, however, we cannot obtain specific information on the tea harvesting time. Nevertheless, to some extent, the risk assessment of perchlorate in tea has rarely referred for the tea consumers, thus this study can present some reference for the future studies.

4. Conclusions

In this study, the occurrence of perchlorate in six different types of tea samples has been monitored, with a detection frequency in samples up to 100%, continuing the trend observed in other studies. Spatial distribution of perchlorate contamination were explored from 13 major tea producing regions of China. Significant differences were found between different regions, with higher perchlorate concentration in tea samples from central China than that from eastern and western China. Perchlorate concentrations from different tea categories were also evaluated, while there was still conflicting results observed between this study and other studies. Hence, the contamination situation in each region is the leading factor of perchlorate contamination in tea. Finally, a human exposure assessment of perchlorate intake through tea consumption was performed by deterministic and probabilistic risk assessment. CDI for the average tea consumers in a point estimate scenario was 0.0183 μg/kg bw/day, accounting for 2.61% based on the RfD value of perchlorate. On average, it is under the safety threshold. Nevertheless, HQs for perchlorate in high tea consumers must draw attention based on a probabilistic risk assessment, if other dietary exposure pathways are considered.

CRediT authorship contribution statement

Yan Li: Investigation, Writing – original draft, Formal analysis. **Shaohua Li:** Resources, Validation. **Jun Ren:** Software, Data curation. **Jingguang Li:** Resources. **Yunfeng Zhao:** Supervision. **Dawei Chen:** Conceptualization, Supervision, Data curation, Writing – review & editing. **Yongning Wu:** Funding acquisition.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.crfs.2023.100606.

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