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Vegetable *Houttuynia cordata Thunb.* as an important human mercury exposure route in Kaiyang county, Guizhou province, SW China

Qingfeng Wang[a,b,*], Zhonggen Li[a,b], Xinbing Feng[b,c], Xinyu Li[b,c], Dan Wang[a], Guangyi Sun[b,c], Huihui Peng[a]

[a] *Department of Resources and Environment, Zunyi Normal College, Zunyi 563006, P.R. China*
[b] *State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, 550002, PR China*
[c] *University of Chinese Academy of Sciences, Beijing, 100049, China*

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ABSTRACT

Consumption of mercury (Hg) contaminated vegetable is one important pathway of Hg expose to humans. In this study, Hg contents in a popular vegetable, *Houttuynia cordata Thunb.* (*HCT*), and its growing soils in Kaiyang county in Guizhou province of southwest China were investigated. Health risk related to Hg exposure through consumption of this vegetable was evaluated for the first time. Hg contents in *HCT* were found to be much higher in three towns in western Kaiyang county (42.3 ± 48.2 μg/kg, FW), where former Hg mines located, than that in other towns (7.6 ± 5.0 μg/kg, FW). Hg contents in *HCT* were also higher than in the other five vegetables (Chinese cabbage, Lettuce, Tomato, Carrot and White radish). Consumption of *HCT* may account for 37.4–61.1% of total vegetable Hg intake of local people in Kaiyang county. Hg concentration in *HCT* positively correlated with that in soil ($r^2 = 0.311$, $p < 0.01$), especially, the labile Hg species (F_{C1}, $r^2 = 0.796$, $p < 0.01$) and the elemental Hg that is bound to the crystalline oxides (F_{C3}, $r^2 = 0.711$, $p < 0.01$), and negatively correlated with Hg that is bound to humic and fulvic complexes (F_{C2}, - 0.304). Estimated daily intake (EDI) and target hazard quotient (THQ) results shown that Hg expose risk is much higher for children than adults, likely due to their different eating habits and the amount of snack intake.

1. Introduction

Since 1956 when the first serious case of mercury (Hg) poisoning disease was found in Minamata, Japan, the toxicity of Hg has been recognized as one of the world's biggest environmental concerns. Mercury is a non-essential element, which can seriously harm human organisms. Mercury can be converted into methylmercury by microorganisms and absorbed and accumulated by crops, posing a great threat to human health through food consumption (Kronberg et al., 2018). Located in the center of the circum-Pacific mercuriferous belt from the perspective of the global plate tectonics, Guizhou province has the largest Hg production in China with at least 12 large and super-large Hg mines in the province. Mercury mining and smelting activities in the past several centuries have resulted in numerous Hg-contaminated sites including Wanshan, Wuchuan and Lanmuchang Hg mining areas (Feng and Qiu, 2008). At present, the quarries of those Hg mining areas have been basically closed, and the tailings piles or dam leftover from Hg mining and smelting are the main secondary sources of Hg pollution. The Hg mining and smelting activities have caused serious Hg pollution to local environment and also posed high health threats to local inhabitants through air inhalation and crop consumption, among different exposure pathways. Rice (Zhao et al., 2019) and vegetables consumption (Jia et al., 2018; Niu et al., 2013) are recognized as the main pathways for Hg expose in inland China (Zhang et al., 2010).

Houttuynia cordata Thunb. (*HCT*) is an extremely popular vegetable in southwest China, especially Guizhou province. The main ways of human's *HCT* intake are raw or stir-fry. *HCT* is normally treated as a root vegetable in Guizhou while as leaf vegetable in other province such as Sichuan and Shanxi in China. It is also well known for its medicinal properties in China as well as other countries, e.g., Japan, Korea, and North-East India. Modern pharmacological experiments show that *HCT* has both anti-inflammatory and anti-oxidative properties (Shingnaisui et al., 2018; Tian et al., 2011).

Nowadays, phytoremediation technology has attracted great attention due to its advantages of cost-effective, nonintrusive, and aesthetically pleasing (Brandao et al., 2018; Lorestani et al., 2013). There are many efforts has been paid to select suitable soil remediation plants

* Corresponding author. Department of Resources and Environment, Zunyi Normal College, Zunyi, 563006, PR China.
E-mail address: wangqingfeng@mail.gyig.ac.cn (Q. Wang).

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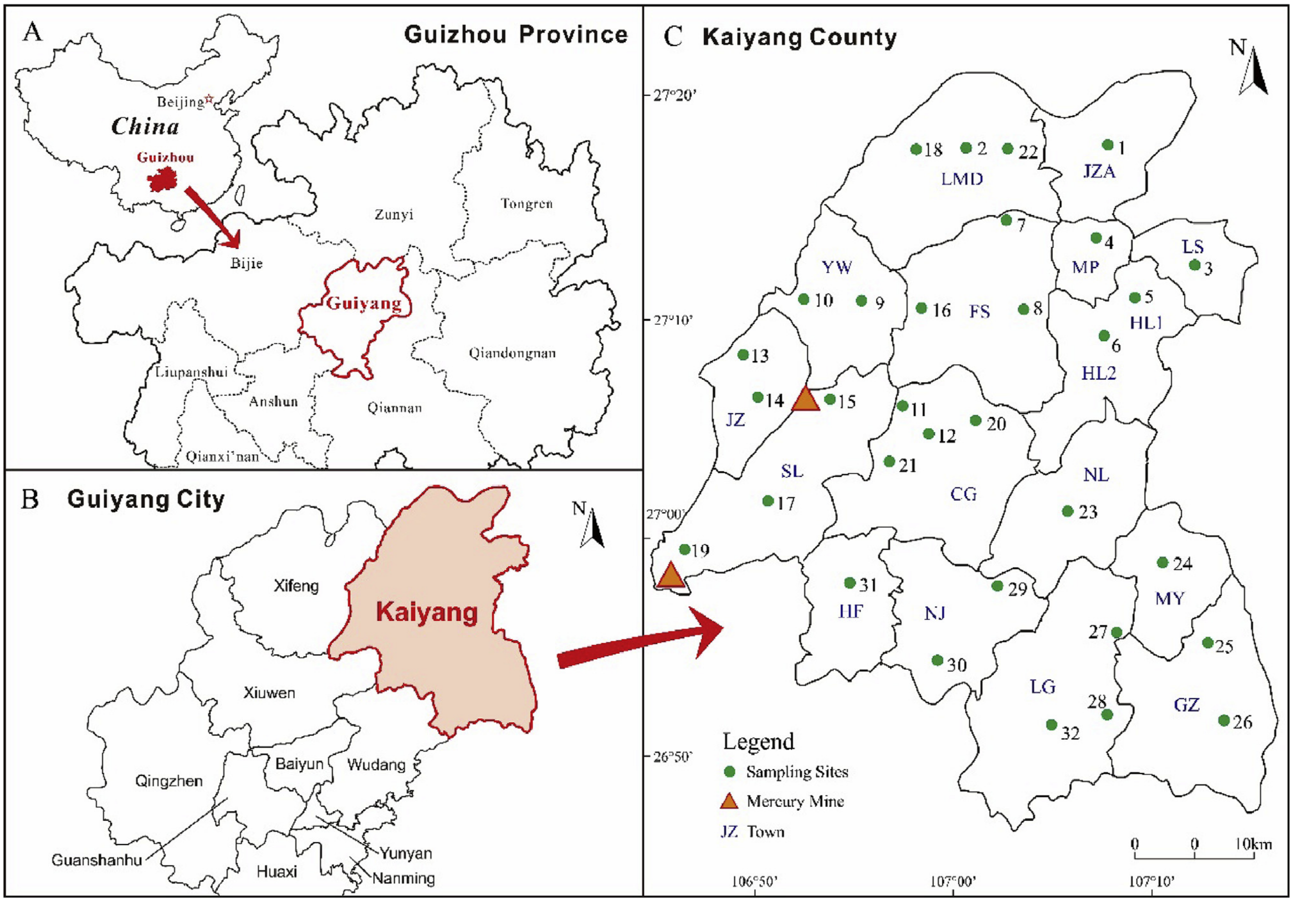

Fig. 1. Map of Guizhou province with sampling locations and historical Hg mining sites.

(Cheraghi et al., 2011; Rugh et al., 1998). *HCT* has a strong heavy metal enrichment capacity (Ha et al., 2011; Liang et al., 2018; Wu et al., 2009), and it could count as an alternative species for remediation of soil contaminated by heavy metals. Wu et al. (2009)showed that *HCT* had a strong adsorption and enrichment effect on Pb, and the content of Pb in leaves was much higher than that in roots and stems. Ha, N. T. H., et al. (Ha et al., 2011) investigated soil phytoremediation and phytometallurgical species in zinc-lead mining areas in Vietnam, and found for the first time that *HCT* is a super-accumulator species of As, and is thus a good candidate for soil phytoremediation of As and other metals. In addition, *HCT* also has a strong enrichment effect on Cd and Cu(Wu et al., 2012; Zhou et al., 2011).

Kaiyang county is part of Guiyang, the capital city of Guizhou province. The famous Kaiyang Hg mining is located in this county with Hg mines mainly distributed in the west area of the county including Shuangliu town (SL), Jinzhong town (JZ) and Yongwen town (YW) (Fig. 1). Besides, Jinzhong town is also one of the three famous phosphate mining areas in China with high-quality rich phosphate ore reserves of about 392 million tons. The exploitation of phosphate ore also causes serious pollution to the local environment (Yang et al., 2015). Guizhou is an important agricultural province in China, and the long history of Hg-mining activities has resulted in serious Hg pollution to the local environment and may bring a greater risk of Hg exposure to agricultural products. *HCT*, used to be a wild vegetable, is now widely planted by farmers in Guizhou province. The main objective of this study was to investigate the Hg concentration levels in the edible part of *HCT* in Kaiyang county and the influencing factors, as well as the associated human health risks through consumption of this vegetable.

2. Material and methods

2.1. Study area and sampling sites

The investigated area chosen in the present study is the whole Kaiyang county located in the northwest of Guiyang city (Fig. 1) in the heart of Guizhou province. The county has an area of approximately 2026 km^2 and a population of 414 000 inhabitants. This region is a mountainous, karstic landscape with an average altitude of 1195 m above sea level. There are more than 500 Hg mine relics in this county, and the mining and Hg production ranked first in the world in the Qing dynasty (A.D.1636–1912) of China. All the Hg mining were closed in the 1980s and 1990s. A total of 32 sites were selected for sampling *HCT* and its growing soils (0–15 cm depth) (Fig. 1). The sampling sites are roughly evenly distributed into each township based on the local population (Table S1).

2.2. Sample collection and total Hg analysis

32 pairs of *HCT* and soil samples, one at each sampling site, were collected in February 2019. Other vegetables (Chinese cabbage, Lettuce, Tomato, Carrot and White radish) were also collected at or around the same sites. All the samples were collected from vegetable patches of local people or wild fields. Only edible parts of *HCT* and other vegetables were kept in the samples and those of *HCT* were the newly growth (< about 2 months) underground tender stems (rhizomes) which have a crisp and refreshing taste. All samples were packed into a polyethylene plastic bag, sealed, identified and stored in a cooler at 4 °C prior to the laboratory analysis. Approximate 0.5 kg of *HCT* or other vegetable and 1 kg of soil were collected at each site. In the laboratory, *HCT* samples were carefully washed and rinsed with

deionized water. The fresh weight was recorded after the surface moisture was dried. Then, the *HCT* samples were placed in nylon mesh bags and hung in a ventilated and dry place to air dried. The dried samples were weighted before being crushed for later use (< 0.150 mm). Soil samples were dried at 40 °C for 7 days before being ground into small pieces (< 0.150 mm). Hg concentrations in *HCT* samples and soils were analyzed by Milestone™ Direct Mercury Analyzer (Model DMA 80, AMA 254 Software) according to EPA method 7473. Analyses of standard reference material (SRM, GBW07405, GSS-5) was used to ensure the quality control and the bias between the recommended value (290 ng/g) and the determined value was less than 5%. SRM were analyzed every five samples to check for the instrument drift, and each sample were analyzed three times to obtain an average value.

2.3. Determination of Hg species in soils

Hg species in soil samples were identified by the CIEMAT sequential extraction method, which was proposed by the Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT, Spain) and could provide more accurate results related to Hg bioavailability than did other existing methods (Dong et al., 2019; Fernández-Martínez and Rucandio, 2013). According to the CIEMAT method, Hg speciation in soils were divided into four forms, namely, labile Hg species (Fc_1), Hg bound to humic and fulvic complexes (Fc_2), elemental Hg and crystalline oxides bound Hg (Fc_3), and Hg sulfide and refractory species (Fc_4). The detailed extraction procedure was reported by Fernández-Martínez, R. (Fernández-Martínez and Rucandio, 2013) and listed in Table S2. Hg in extracting solutions of each step was also measured by DMA 80 and Hg standard stock ($HgCl_2$,1 mg/mL) was provided by national standard substances center of China.

As for reagents, nitric acid (65 percent) and hydrochloric acid (37.5 percent) were obtained from Aladdin reagent (Shanghai) Co. Ltd. All solid reagents used in the sequential extraction were obtained from Sinopharm Chemical Reagent Co., Ltd. Nitric acid and hydrochloric acid were guaranteed reagents and other reagents were analytical reagents.

2.4. Bio-accumulation and health risk assessment methods

Bio-accumulation factor (BAF) is an index of the ability of vegetable to accumulate a particular metal with respect to its concentration in the soil substrate. The health risk assessment method of the edible part of *HCT* was based on that described in Zhuang et al. (2009). The estimated daily intake (EDI) of a particular metal is calculated based on daily food consumption and related metal concentration in food. The target hazard quotient (THQ) is a ratio of determined dose of a pollutant to a reference value. If the ratio is less than 1, the exposed population is unlikely to experience obvious adverse effects. The method of estimating risk using THQ was provided in the U.S. EPA Region III risk-based concentration table (USEPA, 2007). The BAF, EDI and THQ were calculated as follows:

$$BAF = \frac{C_{HC}}{C_{soil}}$$

where C_{HCT} (on dry weight basis, DW) and C_{soil} represent Hg concentration in the edible part of *HCT* and soils, respectively.

$$EDI = \frac{C_{Hg} \times w_{HGT}}{B_w}$$

where C_{Hg} (μg kg^{-1}, on fresh weight basis) is the concentration of Hg in the edible part of *HCT*, W_{HCT} represents the daily average consumption of *HCT* in this region, and B_w is the body weight. Based on the dietary intake survey we conducted, the local inhabitants in Kaiyang had an average *HCT* consumption rate of 76 g day^{-1} per adult with a 55.9 kg body weight and 59 g day^{-1} per child with a body weight of 32.7 kg. Totally 609 local residents (456 adults and 153 children) were involved in the survey and the questionnaire was shown in Table S3. The survey result of adult is much higher than that conducted by the National risk assessment center of China with 5739 respondents of the average dietary consumption amount of the medicine and food homologous herbs (43 g, *HCT* is an important medicine and food homologous herbs) (Zuo et al., 2019). Moreover, as raw materials of Chinese medicine, the medicines containing *HCT* could account for 0.85% of the total 1062 kinds of Chinese medicines in Pharmacopoeia of the People's Republic of China (2010) (Tong Wen et al., 2018).

$$THQ = \frac{EFr \times ED \times FI \times MC}{RfDo \times BW \times AT} \times 10^{-3}$$

where EFr is exposure frequency (365 days $year^{-1}$), ED is exposure duration (76.3years), FI is food ingestion (g $person^{-1}$ d^{-1}), MC is Hg concentration in *HCT* (μg g^{-1}, on fresh weight basis), RfDo is the oral reference dose (μg kg^{-1} d^{-1}), BW is the average body weight (55.9 kg for adult), and AT is averaging time for noncarcinogens (AT = 365 × ED). The oral reference dose of mercury is 0.7 μg kg^{-1} d^{-1} (USEPA, 2009).

2.5. Statistical analysis

Statistical analysis was performed using SPSS 22.0 software and graphs of Hg contents were plotted using Origin (Version 8.5). Relationships between covariant sets of data were subject to regression analysis. Correlation coefficient (r) and significance probability (p) were computed for the linear regression fit. Differences are declared as significant in cases of $p < 0.05$.

3. Results and discussion

3.1. Hg concentration in HCT

HCT was considered to be a heavy metal enrichment plant in many earlier studies (Ha et al., 2011; Wu et al., 2009, 2012; Zhou et al., 2011). Hg contents in *HCT* obtained from the present study are shown in Fig. 2.

Hg concentration in edible part of *HCT* ranged from 2.40 to 145.3 μg/kg with an average of 15.19 ± 26.1 μg/kg on fresh weight basis (FW, moisture content was 82.2 ± 3.5%). 34.3% of samples exceeded the national guidance limit for vegetables, which is 10 μg/kg (FW) recommended by the Chinese National Standard Agency (NHFPC and CFDA, 2017), and the highest Hg concentration in *HCT* was 14.5 times higher than the allowed level, indicating serious Hg contamination in some samples. Large spatial variations were found in Hg concentration of *HCT* with the highest values (42.33 ± 48.2 μg/kg) mainly distributed in the three western towns in Kaiyang county including Yongwen (YW), Jinzhong (JZ) and Shuangliu (SL), while values in the other towns in Kaiyang county were around 7.6 ± 5.0 μg/kg (Fig. 2b and Table S4).

There are only several studies reporting the Hg content in eatable parts of *HCT* and the comparison between different studies were shown in Table S5. It could clearly see that our results were comparable to the study conducted by Zhao et al. (2009) that *HCT* samples collected from several local markets in Wanshan mercury mine. While, this results were much lower than that obtained from the Hg seriously contaminated sites, such as from a wasteland composed of Hg mine tailings in Wanshan mercury mine (Qian et al., 2018) and much higher than other studies (Chen and Wu, 2009; Li et al, 2016, Li et al., 2018b). This indicated that *HCT* was moderately contaminated with mercury by historic mercury mining and smelting activities in Kaiyang county.

Hg concentrations also varied substantially with vegetable species. Hg concentrations tend to be higher in leafy vegetables than root vegetables as the fomer are exposed to atmosphere and accumulate atmospheric Hg^0 through the foliage (De Temmerman et al., 2009; Li

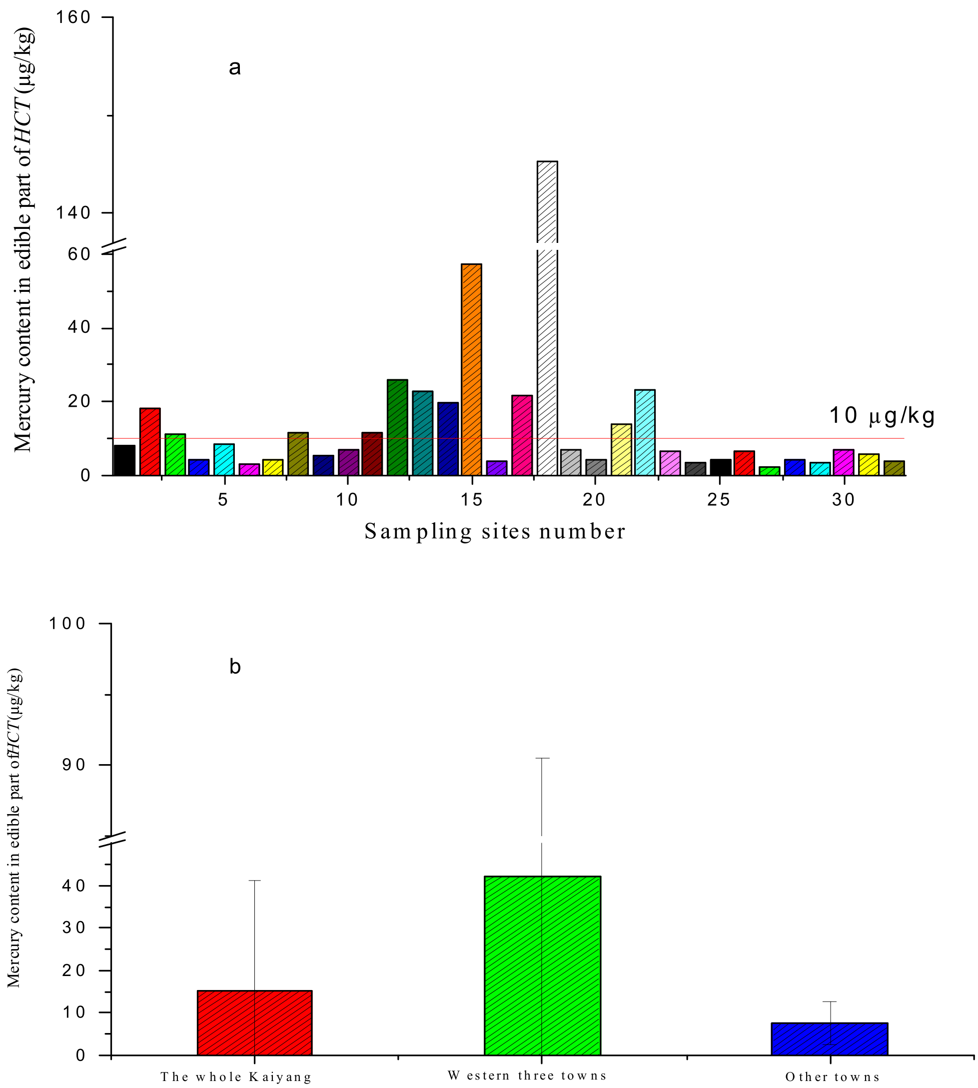

Fig. 2. Hg concentration in the edible part of *HCT* samples a. Each sample sites b. Different area.

et al., 2018a; Shahid et al., 2017). Such a result was also confirmed by several existing studies (Zheng et al., 2018; Yu et al., 2018) which showed that leaf vegetables had the higher Hg content than other vegetables. The comparison of Hg contents in *HCT* and other common vegetables including Chinese cabbage (CC), Lettuce (LT), Tomatoes (TM), Carrot (CR) and White radish (WR) is shown in Table S6 and Fig. 3. It can be seen that *HCT* was the vegetable with the highest Hg content in Kaiyang county, its mercury content was higher than other leaf vegetables such as Chinese cabbage and lettuce. Hg concentration decreased in the order of *HCT* > CC > LT > TM > CR > WR (Fig. 3). It has been commonly accepted that consumption of vegetables and rice is the main exposure of Hg in inland China (Jia et al., 2018; Zhang et al., 2010) and the high Hg concentration in *HCT* may pose a great risk to local people. The *HCT* intake could account for 37.4–61.1% of daily vegetable Hg intake for local people in Kaiyang county according to the follow equation (1) and the calculation result shown in Table 1.

$$\eta = \frac{C_{HCT} \times W_{HCT}}{C_{AV} \times W_{veg}} \times 100\% \quad (1)$$

where η (%) is the ratio of the daily THg intake of *HCT* to that of vegetables. C_{HCT} is the average Hg concentration in *HCT* and C_{AV} is that averaged further from the six vegetables (*Houttuynia cordata Thunb.* (*HCT*), *Chinese cabbage* (CC), *Lettuce* (LT), *Tomatoes* (TM), *Carrot* (CR) and *White radish* (WR)). W_{HCT} is the daily *HCT* intake value of 76 (g $person^{-1}$ d^{-1}) for adult based on our survey and WVeg is the daily vegetables intake value and the value is 368 (g $person^{-1}$ d^{-1}) according to a previous study (Zhang et al., 2010).

Table 1 indicates that the *HCT* consumption could account for as high as 61.1% of total vegetable Hg intake in three western towns, while as low as 37.4% in other towns. By comparing with other vegetables collected from different contaminated sites (Table 2), it could see that the Hg concentrations varied widely, and Hg contents of *HCT* in Kaiyang were much lower than those of vegetables grown in mercury mine (130–2188 μg/kg, FW) (Zhang et al., 2010), but higher than those of the vegetables grown in the other places in Guizhou such as Leigong (4.0 μg/kg, FW), Qingzhen (2.5 μg/kg, FW) and Weining (2.5 μg/kg, FW) (Zhang et al., 2010), as well as higher than those of the samples from some Pb/Zn smelters (< 5 μg/kg, FW) (Li et al., 2018a; Zheng et al., 2007) and (3.5 μg/kg, FW) Chlor-alkali plants (Gibicar et al., 2009).

3.2. Mercury content in soil

Hg concentrations in soil ranged from 59.7 to 2237.0 μg/kg with an average of 519.8 ± 522.9 μg/kg (Fig. S1a) and the geographical variations in Hg contents of soil in different regions of Kaiyang county were similar to those of *HCT* with the highest concentrations in the

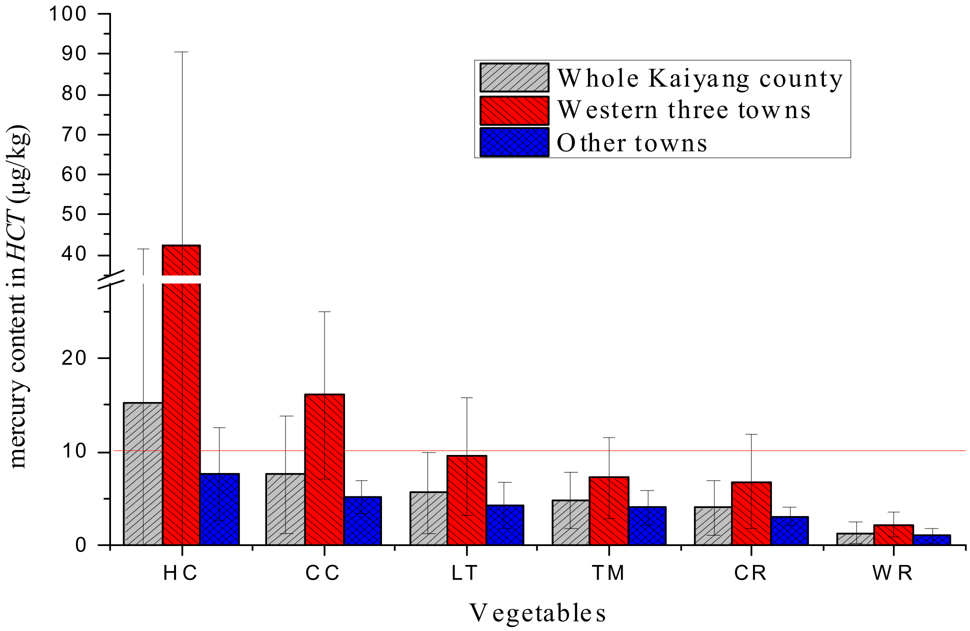

Fig. 3. Average Hg concentration in different vegetables in Kaiyang county (*HCT*: Houttuynia cordata Thunb.; CC: Chinese cabbage; LT: Lettuce; TM: Tomato; CR: Carrot; WR: White radish).

Table 1
Daily vegetable and related Hg intake by local people in Kaiyang county.

Study Area	Vegetable	Mercury content μg/kg, FW	Intake rate (g/d.person)	Share of *HCT* consumption in total vegetable Hg intake (%)
Whole Kaiyang	*HCT*	15.2 ± 26.1	76	49.1
	All six vegetables	6.4 ± 11.9	368	
Western three towns	*HCT*	42.3 ± 48.2	76	61.1
	All six vegetables	14.3 ± 23.3	368	
Other towns	*HCT*	7.6 ± 5.0	76	37.4
	All six vegetables	4.2 ± 3.3	368	

three western towns (Fig. S1b). The mercury concentration in western three towns could be as high as 907.1 ± 630.0 μg/kg.

Compared with the "Soil environmental quality: risk control standard for soil contamination of agricultural land" (MEEC, 2018), 34.38% of the sites exceeded the risk screening value (0.5 mg/kg) and this may indicate that the soil mercury pollution in Kaiyang county has much less than other heavily polluted areas in Guizhou province such as Wanshan and Wuchuan mercury mining area, where soil Hg reached up to tens to hundreds of mg/kg (Feng and Qiu, 2008; Li et al., 2009, 2012).

3.3. Relationship between Hg in HCT and the growing soils

Soil-to-plant transfer is one key component of human exposure to metals through the food chain. The bioavailability of metals in soil depends on the biochemical, chemical and environmental parameters (Natasha et al., 2019). Also, uptake of metals by plants are associated with plant genetic, competition between metals, environmental factors,

Table 2
Comparison of Hg concentration in different vegetables growing in different contamination circumstance.

Vegetables	Mercury content (μg/kg)				Pollutant source	References
	Min.	Max.	Mean	Weight bases		
HCT	2.40 16.59	145.3 666.2	15.2 ± 26.1 81.4 ± 121.2	FW DW	Mercury mining area	This study
Chinese white cabbage, White Radish, Cauliflower, Chinese cabbage, Carrot, Leek, Cabbage, Celery, Garlic sprout and Lettuce	1	11	5	FW	Pb/Zn smelter	Li et al., 2018a,
Aubergine, Chinese cabbage, Leek, Cowpea, Tomato, Allium, Spinach and Pumpkin	0.5	15	4	FW	Pb/Zn smelter	Zheng et al. (2007)
Chinese mustard, radish leaves, Chinese cabbage, and carrot	–	–	130	FW	Mercury mining area	Zhang et al. (2010)
Chinese mustard, radish leaves, Chinese cabbage, and carrot	5	1890	346 ± 471	FW	Mercury mining area	Feng et al. (2008)
cabbage *Chicory parsely*	53 86 125	627 12713 760	215, 280 762, 2188 510, 376	DW	Mercury mining area	Miklavčič et al. (2013)
celery, salad, fennel, *tomato, beet, basil, radish, parsley, aubergines, garlic, onion, bean, celery, gourd, potato*	0.01 0.05	24 111	3.46 ± 4.78 19.57 ± 20.5	FW DW	chlor-alkali plant	Gibicar et al. (2009)

and barriers in plant tissues (Guala et al., 2010; Maria Rosaria et al., 2009). *HCT*, as a root vegetable, usually grows in shade and moist places. Knowledge of the relationship between Hg in *HCT* and Hg in the growing soils is needed designing agricultural planning. BAF value is shown in Fig. S2a. There were significant differences in BAF values among the different sampling sites and the BAF values of *HCT* ranged from 0.012 to 0.510 with an average of 0.20. In an earlier study (Qian et al., 2018), BAF values of the 259 wild plants in wastelands in Wanshan mercury mining area ranged from 0.00028 to 5.5, with values of eight species exceeding 1.0. Despite the lower Hg contents in *HCT* in the present study than those in some other vegetables in the mining area, the high amounts of daily consumption of *HCT* makes it a potential important source of Hg exposure to humans. Though there was no statistically significant difference of BAF value between *HCT* and Chinese cabbage (0.14 ± 0.13) ($p > 0.05$), the average BAF value of *HCT* (0.197 ± 0.132) was indeed significantly higher ($p < 0.05$) than those of the other vegetables, e. g 0.11 ± 0.11 for LT, 0.10 ± 0.11 for TM, 0.08 ± 0.09 for CR and 0.03 ± 0.03 for WR (Fig. S2b).

The relationship between Hg concentration in *HCT* and its growing soils is shown in Fig. S3. A significant positive correlation ($r^2 = 0.311$, $p < 0.01$) was observed, indicating the growing soil determined the amount of Hg in the stem of *HCT* to some extent. On the other hand, the Hg content in soil may not be the only crucial factor, e.g. the case of sample #23. The Hg content in soil of sample 23# was the highest while the Hg in *HCT* was only 26.7 μg/kg (DW) and with the BAF value of only 0.012. Therefore, the relationship between Hg speciation in soil and Hg content in *HCT* was likely affected by multiple factors besides soil Hg content.

Hg speciation in soils increased in the order of F_{C1} (1.09 ± 0.68%) < F_{C3} (13.54 ± 6.43%) < F_{C2} (26.72 ± 4.29%) < F_{C4} (58.66 ± 8.51%)(Fig. S4, Table S7), consistent with previous findings (Fernã and Rucandio, 2014). And Fig. S5 indicates that all Hg fractions in soil show positive correlations with Hg in *HCT*. Intriguingly, results shown in Fig. S5 and Table 3 indicates that both F_{C1} and F_{C3} have high positive correlations with Hg in *HCT*. According to Fernández-Martínez and Rucandio (Fernández-Martínez and Rucandio, 2013), F_{C1} is the labile Hg species and F_{C3} is the elemental Hg and Hg bound to the crystalline oxides. F_{C1} constitutes the most mobile and available Hg fraction including weakly sorbed, weakly ligands forming and highly soluble Hg complexes, and can been easily absorbed by *HCT*. F_{C3} is mainly the elemental Hg in soils and is related to the large variety of historical uses of elemental Hg and industrial and manufacturing sources. F_{C3} significantly correlates with Hg content in plants, as was found in a previous study (Dong et al., 2019). F_{C2} is the fraction of Hg bound to the humic and fulvic complexes, which are highly stable and can be retained in the matrices over long time periods (Fernández-Martínez and Rucandio, 2013). Due to the high affinity of Hg(II) to organic matter (Haitzer et al, 2002), humic and fulvic complexes usually constitute one of the most significant Hg fraction in soils and sediments and, in some cases, can control the behavior of Hg in certain soils and sewage sludges (Biester et al., 2002; Giulio and Ryan, 1987). The affinity of Hg(II) to organic matter may limit the mobility and activity of Hg (Ndungu et al., 2016). Therefore, the correlation between Hg in *HCT* and FC_2 is relatively low. F_{C4} is Hg sulfide and refractory species, which are very stable due to their extremely insolubility ($K_s = 10^{36.8}$ at 20 °C)(Hall et al., 2005). Therefore, the correlation of F_{C4} with Hg concentration in *HCT* is also relatively low. Note that the fractions of Hg in soil often depend on each other, the Pearson correlation analysis might ignore the interactions among different fractions and could not effectively reveal the relationships between the Hg fractions and Hg in *HCT*. Therefore, partial correlation analysis was performed (Table 3). It could be seen that Fc_2 clearly presented a negative partial correlation with Hg content in *HCT*. This result indicates that Hg bound to humic and fulvic complexes (F_{C2}) limit the Hg assimilation, and agricultural activities such as application of farmyard manure and biomass return to field could reduce the Hg concentration in *HCT* to some extent.

Table 4
EDI and THQ of mercury for local residents via *HCT* consumption.

Index	Study area	Population	Mean value
EDI (μg/kg/day)	Whole Kaiyang county	Adult	0.021 ± 0.035
		Child	0.027 ± 0.047
	Western three towns	Adult	0.058 ± 0.065
		Child	0.076 ± 0.087
	Other towns	Adult	0.010 ± 0.007
		Child	0.014 ± 0.009
RfD (μg/kg/day)		All people	0.7
THQ	Whole Kaiyang county	Adult	0.030 ± 0.051
		Child	0.039 ± 0.067
	Western three towns	Adult	0.082 ± 0.094
		Child	0.109 ± 0.124
	Other towns	Adult	0.015 ± 0.010
		Child	0.020 ± 0.013

3.4. Mercury exposure risk from the dietary route

To evaluate the health risk associated with the consumption of *HCT* for the local residents, the dietary intake (EDI) and target hazard quotients (THQ) were calculated (Table 4). EDI and THQ values of the three western towns were much higher than those of the other towns in Kaiyang county, and the values of child were higher than those of adult despite lower amounts of *HCT* consumption by child than adult, which can be mainly explained by their different body weights and ingestion rates of *HCT*. It is also noted that *HCT* is an important ingredient of almost all kinds of snacks in Guizhou, which are consumed more by children than adults.

4. Conclusions

From this study, we obtained a better knowledge regarding the potential risk to human health with respect to the consumption of *HCT*. The intake of *HCT* could take an important part (37.4–61.09%) of vegetable Hg intake, and about one third of samples exceeded the maximum allowed Hg level set by the Food Safety Standards in China. Hg in *HCT* is closely related to Fc_1 (the labile Hg species) and Fc_3(the elemental Hg and Hg bound to crystalline oxides) in soil and has a negative partial correlation with Fc_2(Hg bound to humic and fulvic complexes). EDI and THQ results showed that the children have a much higher risk of Hg exposure than adults, especially in western three towns in Kaiyang county. Those results improve our understanding in public health risk management in China especially in specific rural areas.

Table 3
Correlation between Hg fraction and Hg content in *HCT* (DW).

Correlation analysis	Pearson coefficients	Partial correlation coefficients
F_{C1} labile mercury species	0.796, $p < 0.05$ N = 32	0.602
F_{C2} mercury bound to humic and fulvic complexes	0.259, $p < 0.05$ N = 32	−0.304
F_{C3} elemental mercury and bound to crystalline oxides	0.711, $p < 0.05$ N = 32	0.490
F_{C4} mercury sulfide and refractory specie	0.261, $p < 0.05$ N = 32	0.249

CRediT authorship contribution statement

Qingfeng Wang: Methodology, Investigation, Data curation, Formal analysis, Writing - original draft. **Zhonggen Li:** Conceptualization, Writing - review & editing. **Xinbing Feng:** Conceptualization, Resources, Supervision. **Xinyu Li:** Software, Visualization. **Dan Wang:** Investigation. **Guangyi Sun:** Investigation. **Huihui Peng:** Investigation.

Declaration of comppeting interest

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of the manuscript entitled "Vegetable *Houttuynia cordata Thunb.* as an important human mercury exposure route in Kaiyang county, Guizhou province, SW China".

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecoenv.2020.110575.

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