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# Cadmium exposure as a key risk factor for residents in a world large-scale barite mining district, southwestern China

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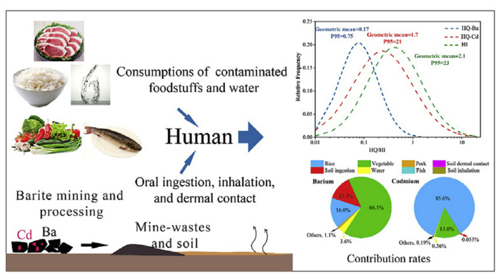
## HIGHLIGHTS

- Exposure risks of coexisting Ba and Cd from multiple exposure pathways at an active Ba mine were evaluated.
- The major pathway for human exposure to Ba was from consumption of vegetables, and that for Cd was from consumption of rice.
- There were unacceptable non-cancer risks to residents from Cd rather than Ba.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Cadmium (Cd) contamination is easily generated during the mining and manufacturing of barium (Ba). In this study, concentrations of both Ba and Cd in rice, vegetables, pork, fish, drinking water, and soil samples from an active barite mining district were determined. Daily intakes of Ba and Cd, as well as corresponding health risks, were evaluated. The average total daily exposure doses of Cd were 0.0035 and 0.0012 mg/kg BW/day (geometric mean) in the mining zone (MZ) and the chemical plant zone (PZ), respectively. These values significantly exceed the provisional tolerable monthly intake (25 μg/kg BW/month, equal to 0.00083 mg/kg BW/day). Based on the daily exposure doses, vegetable consumption was the most significant Ba exposure route for residents, contributing around 66.1% of the total exposure. In contrast, rice consumption was the major Cd exposure pathway, accounting for about 85.6% of the total exposure. Although the geometric mean (0.17) and 95th percentile (P95, 0.75) of the total hazard quotient (*HQ*) for Ba were below the acceptable level (1), suggesting that there were no significant health effects caused by Ba exposure, Cd exposure was associated with significant health risks, with the geometric mean of the *HQ* (1.7) and the P95 (21) well above the acceptable limit (1), indicating the unacceptable non-carcinogenic risk of Cd exposure. In summary, high Cd exposure risk, rather than Ba, was observed for populations living in a large-scale active Ba mining area.



## 1. Introduction

Cadmium (Cd) contamination is one of the most critical environmental issues in China. A recent nationwide survey of arable

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soils indicated that about 7.0% of all sampling sites were contaminated with Cd (MEP, 2014). In Southern China, serious Cd contamination was caused by nonferrous metals mining and smelting activities, including copper, lead, and zinc (Cai et al., 2019; Zhao et al., 2015). Barite was found to enrich Cd. For instance, 9.9 mg/kg and 7 mg/kg of Cd have been reported in barite (Crecelius et al., 2007; Trefry et al., 1986). More recently, the Cd concentration in barite ore was found to be as high as 30.9 mg/kg (Shahab et al., 2016). High levels of Cd in barites can result in the significant release of Cd during barium (Ba) mining and manufacturing. Sediments near abandoned barite mines are found to be heavily contaminated by Cd (Adamu et al., 2015). China enriches barite ore resources, and we also observed elevated Cd concentrations as high as 57 mg/kg in barite (unpublished data). In an active Ba mining area in southern China, Cd concentrations in soil and rice were as high as 91 mg/kg and 3.5 mg/kg, respectively (Lu et al., 2019a).

Both Cd and Ba are non-essential elements for biota (Lamb et al., 2013). Cd is classified as a Group 1 carcinogen and is associated with lung and pancreatic cancer (IARC, 1993; Nordberg, 2009). Cd can also accumulate in organisms, particularly in bone and kidney tissue (Moitra et al., 2013). The notorious itai-itai disease that occurred in Japan was caused by high-dose Cd exposure via rice consumption (Järup and Akesson, 2009). Ba exhibits low toxicity, but a high-dose exposure to Ba can also cause a series of health effects, including cardiac or renal failure, pulmonary edema, respiratory paralysis, and gastric and intestinal hemorrhage (Kravchenko et al., 2014). However, little is known about the risks arising from coexisting Cd and Ba exposure at contaminated sites.

The district of the Dahebian barite mine, which is located in Tianzhu County in the eastern part of Guizhou Province, Southwest China, is known as "land of barite" in China. Currently, Ba mining and manufacturing activities are prominent in the region, with large amounts of barite gangue and ore ($BaSO_4$) piles being introduced at mining sites. Our recent investigation found that Ba and Cd concentrations were up to 65,760 mg/kg and 91 mg/kg, respectively, in paddy soils of this Ba mining area, and there was also a significant positive correlation between the concentrations of Cd and Ba in the soil (Lu et al., 2019a, 2019b), indicating coexistence of Ba and Cd contamination in the region.

Human exposure to heavy metals can occur via ingestion, inhalation, and dermal contact with various contribution rates (Du et al., 2019; Ji et al., 2013; Li et al., 2019; Ng et al., 2019; Qu et al., 2012). de Souza et al. (2017) found that unintentional oral ingestion of soils containing cobalt (Co), Ba, and Cd presented the largest risk to human health in an artisanal gold mine. Francova et al. (2020) reported that oral ingestion of soils may contribute to the daily intake of As, Cd, and Pb that exceeds the tolerable daily intake values. Rehman et al. (2018) reported that drinking water was the most important exposure pathway for prevalent health issues in the vicinity of the Sewakht mines, Pakistan. To obtain a more realistic exposure risk, it is important to consider exposure pathways, other than the dietary ingestion of foodstuffs (Guo et al., 2019; Xu et al., 2020). Hence, multi-pathway health risk assessments are necessary in Ba mining–contaminated areas with significant Cd contamination.

The Dahebian barite mining district, the largest barite mine in China, was selected in the present study to investigate human exposure risks from coexisting Ba and Cd in surroundings. The major goals were to: 1) elucidate the distribution of Ba and Cd in foodstuffs and other environmental matrices; 2) evaluate the daily exposure dose and health risk levels of Ba and Cd; and, 3) identify contribution rates of multiple pathways to Ba and Cd exposure. The findings of this study will provide basic data for environmental contamination and health risk assessment, which can be used for the development of future countermeasures.

## 2. Materials and methods

### 2.1. Study area

The Dahebian barite mining district (26°41′–27°09′ N, 108°54′–109°36′ E) is located in Tianzhu County, Eastern Guizhou Province, Southwest China. The population of Tianzhu County was 0.43 million in 2016. The region has a subtropical humid monsoon climate with an annual average precipitation of 1280 mm and an average annual temperature of 16 °C. Tianzhu is the largest Ba mining and most important Ba salt manufacturing area in China. Ba mining activities began in the 1950s in this area. The ore-forming materials are mainly derived from seawater and hydrothermal sedimentation (Hou et al., 2015). During the mining process, large quantities of waste rock and gangues, accompanied by large amounts of barite ore are piled around the Ba mining sites.

### 2.2. Sample collection

#### 2.2.1. Food samples

A field sampling campaign was conducted during the harvest season in 2017 and 2018. The sampling region was divided into three typical zones (Fig. 1).: the mining zone (MZ, heavily impacted by intensive Ba mining), chemical plant zone (PZ, heavily impacted by Ba salt chemical plants), and control zone (CZ, approximately 20 km away from Ba mining sites).

Vegetables ($n = 117$): Edible parts of vegetables were collected from each region. These included 37 leafy vegetables (including Chinese cabbage, shallot, chive, cabbage, white radish leaf, Chinese white cabbage, lettuce, sweet potato leaf, and leek) and 80 non-leafy vegetables (rootstalk and legume vegetables, including beans, cucumber, ginger, cowpea, bitter gourd, pepper, white radish, pumpkin, eggplant, potato, tomato, sweet potato, loofah, lettuce, winter melon, and white melon). All samples were stored in clean polyethylene bags that were labeled in situ, then transported to the laboratory for sample treatment as soon as possible. Samples were washed with tap water, rinsed with ultrapure water, and then cut into small pieces and freeze-dried (EYELA model FDU-1100, Japan). The dried samples were ground through 150 mesh (0.1 mm, IKA-A11, IKA, Germany) for Ba and Cd concentrations analysis.

Rice ($n = 41$): Rice grain (*Oryza sativa L.*) samples were collected and stored in clean polyethylene bags in situ to avoid cross-contamination. In the laboratory, samples were freeze-dried before being de-husked and then separated into hull, bran, and polished sections (white rice). The polished rice was ground through 150 mesh for Ba and Cd measurements.

Pork ($n = 52$): Pork samples were purchased from local markets ($n = 8$) throughout the whole survey area. Samples were kept in a refrigerator at −20 °C until analysis.

Fish ($n = 34$): Similar to pork samples, fish samples were also purchased from local markets throughout the whole survey area. Samples were kept in a refrigerator at −20 °C until analysis.

#### 2.2.2. Non-food environmental media samples

Soil ($n = 25$): Surface soil samples were collected from farmlands in the survey area and were placed in clean polyethylene bags in situ to avoid cross-contamination. In the laboratory, samples were air-dried, ground with a mortar and pestle, and then filtered through a 200-mesh sieve before further analysis.

Drinking water ($n = 29$): Water samples were collected directly from taps of nearby residences and were stored in borosilicate

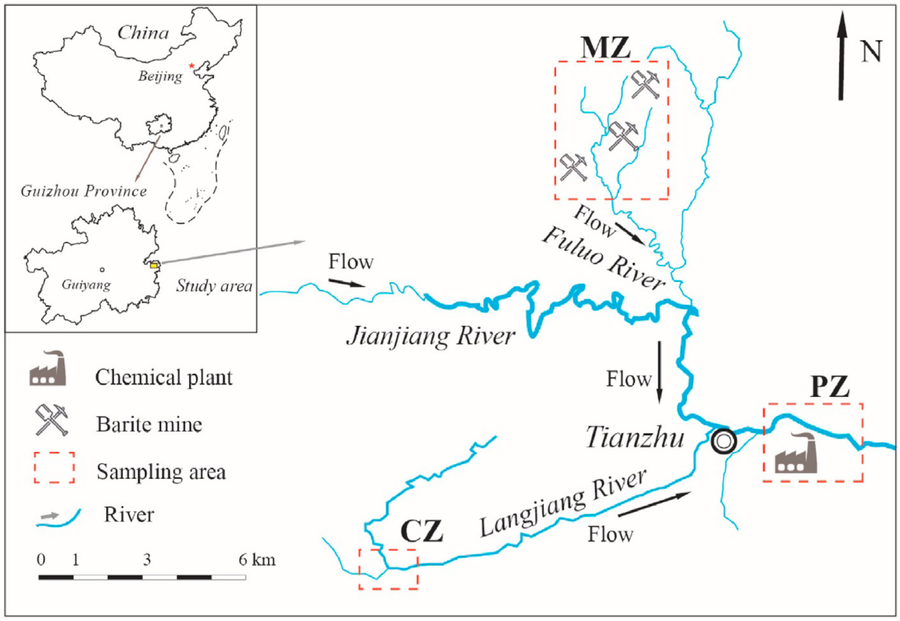


**Fig. 1.** Locations of sampling sites (MZ (the mining zone), heavily impacted by intensive Ba mining; PZ (the chemical plant zone), heavily impacted by Ba salt chemical plants; CZ (control zone), approximately 20 km away from Ba mining sites).

bottles. Water samples were acidified in situ with ultra-pure $HNO_3$ (0.4%, *v*/*v*, Sinopharm Chemical Reagent Company, China) and were then stored in a cool box for transportation to the laboratory. All samples were kept in a refrigerator at 4 °C until analysis.

### 2.3. Sample analysis

Vegetable, rice, pork, and fish samples were digested with $HNO_3/H_2O_2$ (Chen et al., 2013; Lu et al., 2019a). Weighed samples (0.2 g, dry weight for vegetable and rice samples; 0.5 g, fresh weight for pork and fish samples) were placed into a Teflon crucible with $HNO_3$, and the mixture heated in an oven at 150 °C for 36 h $H_2O_2$ was then added to each sample and the samples were heated on a hot plate at 115 °C until complete evaporation of the solution occurred. Deionized water (3 mL) and $HNO_3$ (2 mL) were added to the Teflon crucible, and the mixture maintained at 150 °C for another 12 h.

Soil samples (50 mg, dry weight) were digested with HF and $HNO_3$. The detailed procedure is described in our previous study (Lu et al., 2019a). Briefly, weighed samples were digested with HF and $HNO_3$ in an oven at 160 °C for 48 h. Then, $HNO_3$ (1 mL) was added and the mixture heated on a hot plate at 115 °C until complete evaporation of the solution occurred. Then, deionized water (3 mL) and $HNO_3$ (2 mL) were added to dissolve any residues.

Concentrations of Cd and Ba in all samples were determined by inductively coupled plasma mass spectrometry (ICP-MS; Agilent HPLC 1290-7700x, USA), except for soil Ba, which was measured by inductively coupled plasma atomic emission spectroscopy (ICP-OES; Wasst-mpx, USA).

Residents in this area primarily consume fresh vegetables, and guidance values for heavy metals in vegetables in China are based on their fresh weights. Therefore, concentrations of both Ba and Cd in vegetable samples were converted to fresh weight concentrations based on the reported moisture contents of the different vegetable types according to the United States Environmental Protection Agency (USEPA 1997). Similarly, Ba and Cd in meat samples were directly recorded as fresh weight concentrations.

Duplicates, method blanks, and certified reference materials (GBW07405, yellow-red soil; GBW10020, citrus leaf; GBW09101B, human hair) were employed for quality assurance and quality control (QA/QC) in the Ba and Cd concentration analyses. Detailed information is provided in Table S1.

During the analysis process for all samples, diluted standard stock solutions were measured every 20 samples, with the recoveries of Ba and Cd ranging from 95% to 106% and 90%−108%, respectively. The detection limit of ICP-MS is 0.4 ng/L for Cd and Ba, and the detection limit of ICP-OES is 5 μg/L for Ba.

### 2.4. Health risk assessment

#### 2.4.1. Average daily exposure dose

In the present study, the exposure risks of oral ingestion (including food consumption, water drinking, and soil ingestion), inhalation, and dermal contact were considered. The dose obtained from contact with soil (including ingestion, inhalation, and dermal contact) was calculated according to following equations (USEPA, 2001; Li et al., 2015):

$$ADD_{ing} = \frac{CS \times IRing \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{1-1}$$

$$ADD_{inh} = \frac{CS \times IR_{inh} \times EF \times ED}{PEF \times BW \times AT} \tag{1 −2}$$

$$ADD_{dermal} = \frac{CS \times SA \times SL \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{1 −3}$$

While the dose through ingestion of food and water was calculated as follows:

$$ADD_{food/water} = \frac{C \times IR_{food/water} \times EF \times ED}{BW \times AT} \tag{1 −4}$$

Here, $ADD_{ing}$, $ADD_{inh}$, $ADD_{dermal}$, and $ADD_{food/water}$ represent the average daily dose (mg/kg BW/day) received through soil ingestion, inhalation, dermal contact, and daily intake of food or water, respectively; *CS* is the concentration (mg/kg) of Ba or Cd in soil; and *C* is the Ba or Cd concentration (mg/kg or μg/L) in rice, vegetables, pork, fish, or water. $IR_{ing}$ is the ingestion rate; $IR_{inh}$ is the inhalation rate; $IR_{food/water}$ is the food or water ingestion rate; *EF* is the exposure frequency; *ED* is the exposure duration; *BW* is the body weight; *AT* is the average time for noncarcinogens; *SA* is surface area of the skin; *SL* is the skin adherence factor; and *ABS* is the dermal absorption factor. The values of each exposure parameter in the above equations are summarized in Table S2.

### 2.4.2. Calculation of non-carcinogenic risk

The hazard quotient (*HQ)* is used to assess non-carcinogenic risk. The equations for calculating *HQ* are as follows:

$$\mathrm{HQ_{ing}} = \frac{\mathrm{ADD_{ing}}}{\mathrm{RfD}_{ing}} \tag{2-1}$$

$$HQ_{inh} = \frac{ADD_{inh}}{RfD_{inh}} \tag{2-2}$$

$$HQ_{dermal} = \frac{ADD_{dermal}}{RfD_{dermal}} \tag{2 -3}$$

$$HQ_{food/water} = \frac{ADD_{food/water}}{RfD_{food/water}} \tag{2 -4}$$

Here, $RfD_{ing}$, $RfD_{inh}$, $RfD_{dermal}$, and $RfD_{food/water}$ are $2.0 \times 10^{-1}$, $1.43 \times 10^{-4}$, $4.90 \times 10^{-3}$, and $2.0 \times 10^{-1}$ mg/BW kg/day for Ba (De Miguel et al., 2007; Li et al., 2017b), and $1.00 \times 10^{-3}$, $1.00 \times 10^{-3}$, $1.00 \times 10^{-5}$, and $1.00 \times 10^{-3}$ mg/BW kg/day for Cd (Liu et al., 2019; Li et al., 2017b).

The hazard index (*HI*), described as the sum of the *HQs* of different elements (in this study, the elements assessed were Ba and Cd), was assessed according to the method described in previous research (Sang et al., 2019; Yi et al., 2011). The equation for calculating *HI* is as follows:

$$HI = HQ_1 + HQ_2 + \cdots\cdots + HQ_n \tag{2 -5}$$

Here, 1, 2,…and n represent different elements.

## 2.5. Monte-Carlo simulation

Probabilistic calculations are more reliable than deterministic calculations (Koupaie and Eskicioglu, 2015), in assessing the health risks of heavy metals (Peng et al., 2016; Xu et al., 2020). In the present study, Monte-Carlo simulation was performed using Crystal Ball (version 11.2.4; Oracle Co. Ltd., USA) embedded in Microsoft Excel 2013 (Microsoft Co. Ltd., USA). Based on 100,000 iterations, the average total daily exposure dose (TADD), *HQ, HI,* and the contribution rates of each exposure pathway were simulated.

## 2.6. Statistical analysis

Data were analyzed using SPSS 25. Means were compared through one-way ANOVA. The capital letters represents the significant difference at the 1% level; and the lower letters represents the significant difference at the 5% level.

# 3. Results and discussion

## 3.1. Ba and Cd concentrations

### 3.1.1. Main foodstuffs

*3.1.1.1. Ba concentrations.* The Ba concentrations in food samples collected from the three zones are presented in Table 1 and Fig. 2. The average concentrations of Ba in rice in the MZ (1.0 ± 1.2 mg/kg) and PZ (0.79 ± 0.75 mg/kg) were about three times higher than those in the CZ (0.30 ± 0.17 mg/kg, $p < 0.05$). For vegetables, the average Ba concentrations in the three areas were 4.3 ± 4.5 mg/kg (MZ), 3.3 ± 2.6 mg/kg (PZ), and 2.4 ± 1.6 mg/kg (CZ). As the pork and fish samples were collected from local markets across the three zones, the Ba concentrations in fish and pork were representative of those in the whole region. The average concentrations were 0.47 ± 0.81 mg/kg for pork and 0.69 ± 0.81 mg/kg for fish (Table 1, Fig. 2).

The average Ba concentrations in food samples were in the following order: vegetables>rice>fish>pork (Figure S1a). The Ba concentration in vegetables was about 4.3 times, 4.2 times, and 8 times higher than that in rice in the MZ, PZ, and CZ, respectively ($p < 0.05$). Additionally, leafy vegetables exhibited higher Ba concentrations than non-leafy vegetables ($p < 0.01$). McBride et al. (2014) obtained similar results via investigation of the Ba concentrations in urban garden vegetables. They proposed that the vegetables absorbed Ba via their roots, with minimal Ba transfer to the fruiting parts of the vegetables. Other researchers have also reported that leafy crops can accumulate high levels of Ba, with concentrations reaching 100 mg/kg (dry weight) or even much higher (Lamb et al., 2013; Nabulo et al., 2012). Leafy vegetables appear to have the highest propensity to accumulate Ba, indicating their high translocation and transpiration rates. Non-leafy vegetables transfer metals from the roots to the stem and then to the fruit, which takes longer and therefore results in lower accumulation (Itanna, 2002; Gupta et al., 2019).

*3.1.1.2. Cd concentrations.* The average Cd concentrations in rice were 1.1 ± 1.1, 0.28 ± 0.31, and 0.06 ± 0.04 mg/kg in the MZ, PZ, and CZ, respectively (Table 1 and Fig. 2). About 78% and 44% of rice samples in the MZ and PZ, respectively, exceeded China's permissible rice Cd value (0.2 mg/kg, Ministry of Health of the People's Republic of China, 2017). Cd concentrations in rice obtained from MZ were higher than those obtained from the CZ ($p < 0.05$), suggesting that Ba mining could release Cd into the environment.

For vegetables, the average Cd concentrations in the three areas were 0.099 ± 0.29, 0.08 ± 0.12, and 0.049 ± 0.042 mg/kg, for the MZ, PZ, and CZ, respectively. The maximum permissible Cd values in China are 0.2 mg/kg and 0.1 mg/kg in leafy vegetables and in rootstalk or legume vegetables, respectively (Ministry of Health of the People's Republic of China, 2017). Compared with these maximum permissible values, approximately 9.7% of leafy vegetable samples and 13% of non-leafy vegetable samples exceeded the permissible values at two zones (MZ and PZ). Cd concentrations in all vegetable samples collected from the CZ were lower than the limit values. The average Cd concentrations in pork and fish were 0.0027 ± 0.011 and 0.0019 ± 0.0023 mg/kg, respectively, across the region, both of which were lower than the maximum permissible Cd values for pork and fish (0.1 mg/kg; Ministry of Health of the People's Republic of China, 2017).

The average Cd concentrations in food samples were in the following order: rice > vegetables > pork > fish (Figure S1b). As expected, rice exhibited much higher Cd concentrations than vegetables ($p < 0.01$), which was similar to the results of Song et al. (2017). Rice usually accumulates more Cd than other cereals (Meharg et al., 2013), which can be attributed to the high

**Table 1**
Average and range (in brackets) of concentrations of Ba and Cd in the collected samples from the three zones.

| Sample type | MZ | | PZ | | CZ | |
|---|---|---|---|---|---|---|
| | Ba | Cd | Ba | Cd | Ba | Cd |
| Rice (mg/kg) | 1.0 ± 1.2 (0.10–5.0) | 1.1 ± 1.1 (0.02–3.5) | 0.79 ± 0.75 (0.23–3.5) | 0.28 ± 0.31 (0.01–1.1) | 0.30 ± 0.17 (0.14–0.58) | 0.06 ± 0.04 (0.006–0.11) |
| Leafy vegetable (fresh, mg/kg) | 5.1 ± 3.5 (0.71–13) | 0.11 ± 0.29 (0.02–1.3) | 4.6 ± 3.2 (0.74–11) | 0.14 ± 0.19 (0.01–0.68) | 3.0 ± 0.43 (2.5–3.5) | 0.088 ± 0.036 (0.048–0.14) |
| Non-leafy vegetable (fresh, mg/kg) | 3.9 ± 4.9 (1.12–19) | 0.094 ± 0.29 (0.0057–1.8) | 2.7 ± 2.1 (0.38–9.3) | 0.058 ± 0.067 (0.005–0.29) | 2.03 ± 1.8 (0.52–7.3) | 0.023 ± 0.019 (0.0092–0.063) |
| All vegetable (fresh, mg/kg) | 4.3 ± 4.5 (0.71–19) | 0.099 ± 0.29 (0.0057–1.8) | 3.3 ± 2.6 (0.38–11) | 0.080 ± 0.12 (0.0053–0.68) | 2.4 ± 1.6 (0.52–7.3) | 0.049 ± 0.042 (0.0092–0.14) |
| Pork (fresh, mg/kg) | 0.47 ± 0.81 (0.03–5.1) | 0.0027 ± 0.011 (0.00015–0.084) | 0.47 ± 0.81 (0.03–5.1) | 0.0027 ± 0.011 (0.00015–0.084) | 0.47 ± 0.81 (0.03–5.1) | 0.0027 ± 0.011 (0.00015–0.084) |
| Fish (fresh, mg/kg) | 0.69 ± 0.811 (0.04–2.9) | 0.0019 ± 0.0023 (0.0002–0.0093) | 0.69 ± 0.811 (0.04–2.9) | 0.0019 ± 0.0023 (0.0002–0.0093) | 0.69 ± 0.811 (0.04–2.9) | 0.0019 ± 0.0023 (0.0002–0.0093) |
| Drinking water (μg/L) | 34 ± 30 (7.1–106) | 0.17 ± 0.19 (LD–0.57) | 41 ± 36 (8–115) | LD | 6.5 ± 1.2 (5.6–7.4) | LD |
| Soil (dry, mg/kg) | 12,645 ± 15,124 (816–42,113) | 7.2 ± 13 (0.35–41) | 1811 ± 1452 (563–5313) | 0.47 ± 0.36 (0.11–1.3) | 614 ± 88 (548–744) | 0.22 ± 0.11 (0.11–0.37) |

"LD" is less than the detection limit.

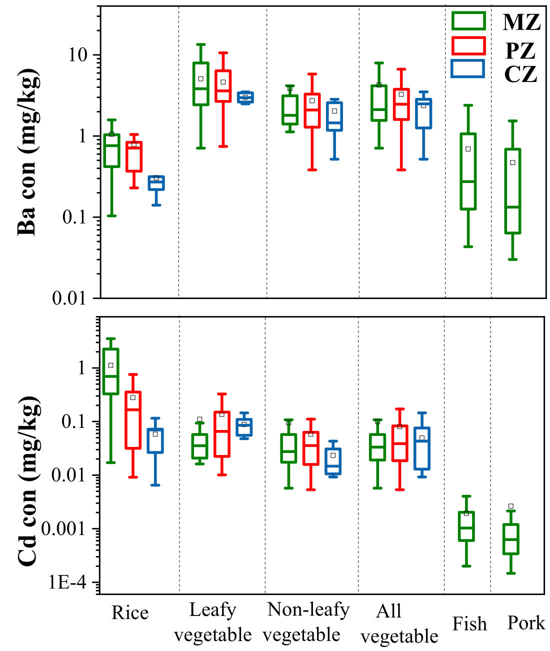


**Fig. 2.** Barium and Cd concentrations in rice, leafy vegetables, non-leafy vegetables, all vegetables, fish, and pork. The black squares represent the mean values. The boxes represent the 25th, 50th, and 75th percentiles.

expression of *OsNramp5* and Nramp5 proteins (Sui et al., 2018). Additionally, the Cd concentration in rice cultivated in the MZ was higher than that of rice from the other two zones ($p < 0.05$). Additionally, we observed a significant positive relationship between Cd concentrations in rhizosphere soils and rice (Lu et al., 2019a); therefore, the elevated Cd concentration in rice is likely to be related to the high Cd concentration in soils.

### 3.1.2. Other matrices

*3.1.2.1. Ba concentrations in drinking water and soil.* The average Ba concentrations in drinking water in the three areas were 34 ± 30, 41 ± 36, and 6.5 ± 1.2 μg/L in the MZ, PZ, and CZ, respectively (Table 1, Figure S2); these were lower than the maximum acceptable Ba concentration in drinking water of 700 μg/L, as recommended by the WHO (2011). However, the average Ba concentrations in drinking water in the MZ and PZ were higher than the reported Ba value of 10 μg/L for surface water in China (Ministry of Environment Protection of China, 2002) and higher than the Ba concentration in drinking water from the CZ. Elevated Ba indicated that the drinking water in these two zones was contaminated by Ba, to a certain degree.

The average Ba concentrations in soils in the three zones were 12,645 ± 15,124, 1811 ± 1,452, and 614 ± 88 mg/kg in the MZ, PZ, and CZ, respectively (Table 1). Elevated soil Ba concentrations obtained from the MZ were 20 times higher than that of soil from the CZ ($p < 0.01$). Similarly, soil Ba concentrations in the PZ were also higher than in the CZ ($p < 0.05$). In a previous investigation of soil around Ba salt chemical plants (Zhang et al., 2012), the average soil Ba concentration was 1666 ± 767 mg/kg, which was similar to our results. The elevated Ba in the soil around the Ba salt chemical plants was attributed to the long-term emission of Ba salt from the chemical plant, which is then transported to the surface of the soil via dry and wet deposition and then migrates and transforms in the soil (Zhang et al., 2012).

*3.1.2.2. Cd concentrations in drinking water and soil.* The Cd

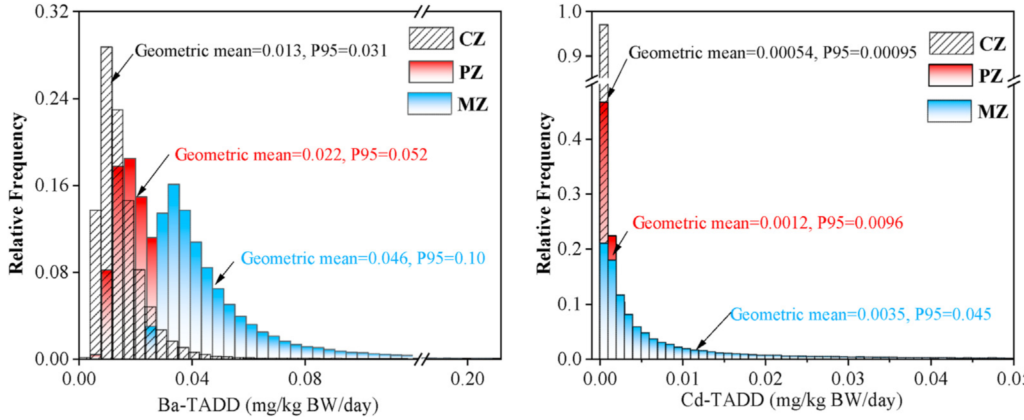


**Fig. 3.** Distribution of the average total daily exposure dose of Ba and Cd in the three zones.

concentrations in drinking water collected from the CZ and PZ were below the detection limit (4 ng/L). The average Cd concentration in drinking water from the MZ was 0.17 ± 0.19 μg/L (Table 1), which was lower than the maximum permissible limit of Cd in drinking water in China (5 μg/L; Ministry of Health of the People's Republic of China and Standardization administration, 2006), and much lower than other reported values for drinking water from other contaminated areas. The Cd concentrations in drinking water from 10 cities in Saudi Arabia, for instance, were reported to range from 2 to 10 μg/L due to the effects of the Gulf War and the Kuwaiti oil fires (Hashem, 1993). In Kamrup district, Assam, India, the Cd concentration of drinking water was also reported to reach up to 25 μg/L as a result of geogenic contamination (Chakrabarty and Sarma, 2011).

For soil samples, the average Cd concentrations were 7.2 ± 13, 0.47 ± 0.36, and 0.22 ± 0.11 mg/kg in the MZ, PZ, and CZ, respectively (Table 1). The average value at MZ was about 33 times higher than at CZ ($p < 0.05$) and approximately 22 times higher than the national Grade II value of 0.3 mg/kg for Cd in soil in China (Ministry of Environment Protection of the People's Republic of China, 1995). Compared with Cd concentrations in other contaminated soils, such as soils from wastewater irrigation areas (mean value, 0.46 ± 0.02 mg/kg), coal mining areas (mean value, 0.3 ± 0.16 mg/kg), and lead-zinc mining districts (mean value, 1.62 ± 1.45 mg/kg) as reported by Wang et al. (2012), Cheng et al. (2018), and Nnabo (2015), respectively, the soil Cd contamination in the MZ in the present study is more significant.

### 3.2. Daily exposure dose

In the present study, the TADDs of Ba and Cd were calculated using the Monte-Carlo simulation technique, with 100,000 iterations. Results for the three zones, with lognormal distributions, are shown in Fig. 3. The geometric mean Ba TADD values were 0.046, 0.022, and 0.013 mg/kg BW/day for the MZ, PZ, and CZ, respectively. The Ba TADD P95 values were 0.1, 0.052, and 0.031 mg/kg BW/day for the MZ, PZ, and CZ, respectively. For Cd, the geometric mean values were 0.0035, 0.0012, and 0.00054 mg/kg BW/day for the MZ, PZ, and CZ, respectively, and the Cd TADD P95 values were 0.045, 0.0096, and 0.00095 mg/kg BW/day for the MZ, PZ, and CZ, respectively. Generally, Ba and Cd exposure risk levels were all higher for the CZ than the other two zones (Fig. 3), suggesting that mining activities are the most important contributors to Ba and Cd contamination.

With respect to the geometric mean Ba TADD values, CZ (0.013 mg/kg BW/day; equivalent to 0.78 mg/d) exhibited a lower exposure value than the total Ba exposure level (1.24 mg/d) reported by Schroeder and Kraemer (1974), but higher exposure levels were observed at MZ and PZ. Compared to 0.53 mg/day, which is the total dietary Ba exposure reported for the population in the United Kingdom (Ysart et al., 1999), the Ba exposure level in the current study was elevated, even at CZ.

The geometric mean values of total Cd exposure doses at MZ, PZ, and CZ were 4.2, 1.4, and 0.64 times higher than the provisional tolerable monthly intake values (25 μg/kg BW/month, equivalent to 0.00083 mg/kg BW/day) established by the JECFA (FAO/WHO, 2010). In particular, at the MZ the Cd TADD was as high as 0.0035 mg/kg BW/day (equivalent to 210 μg/day), which was approximately 6.4 times higher than 32.7 μg/day, which is the average dietary intake of Cd in 20 regions of China (Wei et al., 2019). This result indicated that Ba mining releases a significant quantity of Cd into the environment. Compared with the data obtained from polluted areas of southwest China (Zhu et al., 2016; Huo et al., 2018), the exposure level at the MZ in the present study (105 μg/kg BW/month) was elevated, suggesting that special attention should be paid to the environmental issues concerning Cd contamination in this mining area.

### 3.3. Hazard quotient/hazard index

Monte Carlo simulated *HQ* and *HI* values were calculated for the whole study area and are shown in Fig. 4. Values of *HQ* or *HI* that are greater than 1 indicate high non-carcinogenic risks to human health (Yu et al., 2010). The geometric mean and P95 values of the total *HQ* for Ba were 0.17 and 0.75, respectively, suggesting that Ba exposure may not pose a significant potential health risk to the population in the study area.

However, the geometric mean *HQ* value for Cd was 1.7, above the threshold value of 1, and the P95 was up to 21, greater than the threshold value of 1. This indicated a high potential risk to the health of the population. In the present study, the geometric mean value of 1.7 for adults was comparable to the value of 2.22 for both males and females reported in a Cd-contaminated area of Xiangtan (Chen et al., 2018), but much higher than 0.54, an average *HQ* value across all of China (Wei et al., 2019).

The geometric mean value of *HI* was 2.1 and the P95 was 23. The high *HI* value in the studied Ba mining area suggested that human exposure to Ba and Cd poses an unacceptable non-carcinogenic health risk, which is mainly due to Cd intake. Therefore, there is an urgent need for measures to mitigate Cd contamination in this area.

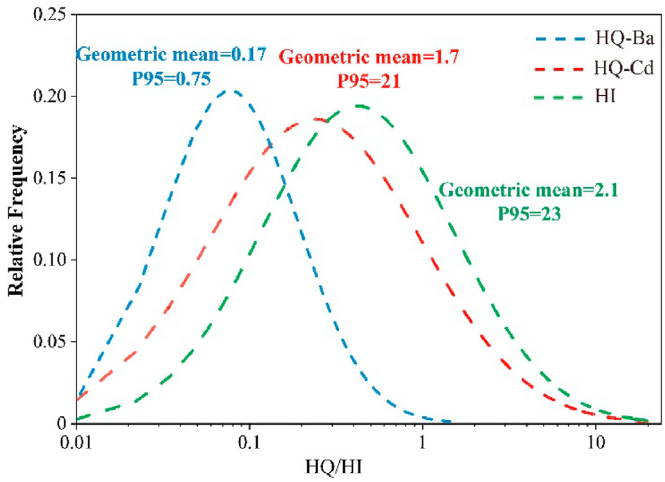


**Fig. 4.** Distributions of the total hazard quotients of Ba and Cd, and the hazard index across the whole study area.

### 3.4. Contribution rates

In the present study, the Ba and Cd contribution ratios of different pathways, across the whole population, were simulated via the Monte-Carlo simulation technique with 100,000 iterations. The median value was obtained to represent the average contribution ratio of each pathway, as shown in Fig. 5.

Ingestion of vegetables accounted for the majority of Ba TADD (66.1%), with little contribution from pork consumption, fish intake, soil dermal contact, and soil inhalation (these four pathways only accounted for 1.1% of the TADD). The Ba contribution ratio of drinking water was 3.6%, which was in contrast to previous results indicating that exposure to Ba occurs mostly from drinking contaminated water (Padilla et al., 2010). Importantly, oral ingestion of soils accounted for 13.2% of the Ba TADD in the whole area, suggesting that exposure doses via soil ingestion cannot be ignored. This finding was similar to the results of previous studies that indicated ingestion of contaminated soil particles as an important heavy metals exposure pathway, resulting in high exposure risks for residents (Li et al., 2017a; Wang et al., 2018; Xu et al., 2020).

For Cd, rice consumption accounted for 85.6% of the TADD, followed by vegetable consumption (13.8%; Fig. 5). The contribution rates of drinking water, consumption of pork and fish, oral ingestion of soil, dermal contact with soil, and inhalation of particles to Cd intake were much lower, with the total contribution ratio from these pathways being less than 1%. A high Cd intake contribution ratio from rice consumption has been widely reported in previous studies: as much as 70–87% of the total Cd exposure dose for the population in South China was reported to be from rice consumption (Zhu et al., 2016; Huo et al., 2018). For the general population in China, rice consumption contributes over 55.8% to the total Cd exposure dose (Song et al., 2017). In other Asian countries, rice consumption is also the major pathway of Cd exposure, accounting for 90% of total Cd exposure dose in Vietnam and 31% in Korea (Kim and Wolt, 2011; Marcussen et al., 2013).

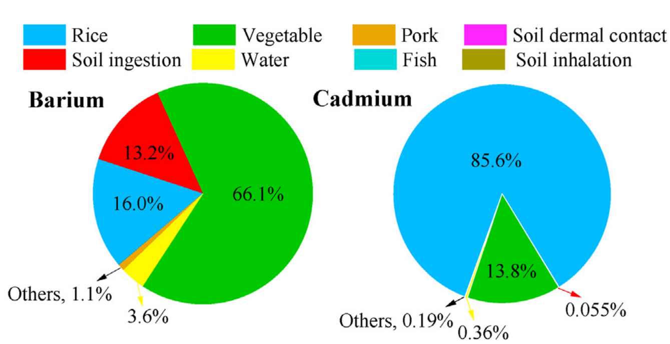


**Fig. 5.** Percentage contribution of each exposure pathway to the total daily exposure doses of Ba and Cd.

### 3.5. Implications

Elevated Cd exposure from the consumption of locally cultivated rice is a significant issue highlighting the requirement to prioritize action and strategies to mitigate the Cd exposure risk in barite mining and manufacturing regions. Hence, implementing measures to reduce human health risks in the present Ba mining area—such as planting low Cd-accumulating rice varieties, reducing the consumption of home-grown rice, or remediating Cd-contaminated paddy soil—would decrease Cd exposure through rice. In addition, the local population could reduce their Ba exposure by reducing the consumption of locally cultivated vegetables, since vegetable ingestion was a major Ba contributor to the risk apportionment.

Generally, different age subgroups have different heavy metals exposure risks (Chen et al., 2018). In this study, only risk levels for the adult population were calculated. Additionally, because data on specific exposure factors for the local population were not available, the exposure risks in this study were calculated based on general exposure factors (consumption of water, rice, fish, pork, and vegetables from previous studies, and soil exposure parameters based on data from the USEPA), which may be unsuitable and lead to unsuitable risk levels. Thus, there may be unacceptable exposure risks for sensitive populations. Previous studies have indicated that children are more susceptible to environmental pollution than adults due to their underdeveloped immune systems (Cao et al., 2015). In the present study, because the elevated Cd exposure was found to pose an unacceptable non-carcinogenic risk for adults, there is no doubt that children will suffer an even greater Cd exposure risk in this area. Despite these limitations, however, the present study provides a comprehensive analysis of Ba and Cd exposure levels in Ba mining areas. Our results provide useful information for the development of future countermeasures in the study area.

## 4. Conclusions

The present study quantified the daily exposure doses and exposure characteristics of Ba and Cd in an active Ba mining area. The results indicated that the geometric mean values of the TADD of Cd in the MZ and PZ were relatively high, corresponding to 4.2 times and 1.4 times the provisional tolerable monthly intake (25 μg/kg BW/month, equal to 0.00083 mg/kg BW/day) established by the JECFA. The population inhabiting the present Ba mining area is exposed to higher levels of Cd than Ba. With respect to the exposure characteristics of Ba and Cd, vegetable consumption was found to be an important route for Ba exposure accounting for 66.1% of the total exposure, but rice intake was the predominant pathway for Cd exposure for the population living in this Ba mining area with contributing about 85.6%. Additionally, we found that the oral ingestion of soil was an important Ba exposure pathway for residents. Overall, elevated Cd exposure risk is a significant issue in the present study area. Future studies must systematically assess Cd exposure risks in other Ba mining areas.

## Author statements

Dr. Qinhui Lu: Conceptualization, Investigation, Methodology, Data process, Writing – original draft, Monte Carlo Model operations, Visualization, Dr. Zhidong Xu: Investigation, Methodology, Visualization, Dr. Xiaohang Xu: Investigation, Methodology, Dr. Lin Liu: Investigation, Dr. Longchao Liang: Investigation, Prof. Zhuo Chen: Supervision, Funding acquisition, Ms. Xian Dong: Resources, Ms. Chan Li: Investigation. Prof. Guangle Qiu: Writing – review & editing, Project administration, Funding acquisition.

## Declaration of competing interest

The authors declare no conflict of interest.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2020.129387.

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