



Spatial distribution of heavy metals in rice grains and human health risk assessment in Hunan Province, China

Hongsheng Cui^{1,2} · Jia Wen^{1,2} · Lisha Yang^{1,2} · Qi Wang^{1,2}

Received: 12 April 2022 / Accepted: 19 June 2022 / Published online: 27 June 2022
© The Author(s), under exclusive licence to Springer-Verlag GmbH Germany, part of Springer Nature 2022

Abstract

Rice is the main food in China, and its pollution by heavy metals has attracted widespread attention. In this study, rice grain samples were collected from 14 prefecture-level cities in Hunan Province, China. The contents of 9 heavy metals (i.e., As, Cr, Co, Ni, Cu, Zn, Cd, Pb, and Sb) were measured using graphite digestion-inductively coupled plasma mass spectrometry (ICP-MS). Pearson correlation analysis and principal component analysis were performed to evaluate the correlation among these heavy metals. In addition, ordinary kriging interpolation were applied to investigate the spatial distribution pattern of the heavy metals. Results showed that the average concentrations of these heavy metals were 0.48 (As), 1.28 (Cr), 0.03 (Co), 0.84 (Ni), 2.39 (Cu), 15.73 (Zn), 0.28 (Cd), 0.66 (Pb), and 0.0043 (Sb) mg/kg, respectively. The single-factor pollution index (SFPI) contamination assessment showed that As, Pb, Cr, Ni, and Cd accumulated significantly in the rice grain, with over-standard rates of 100%, 100%, 64.70%, 47.05%, and 44.12%, respectively. The Sb concentrations at the sampling sites were low, and there was no obvious pollution. Health risk assessment showed that the target hazard quotient followed the order of As > Cr > Cd > Pb > 1.0 > Co > Cu > Zn > Ni > Sb, and the carcinogenic risk value was in the order of Cd > Ni > As > Cr > 1.0×10^{-4} > Pb. In particular, quick actions should be taken to regulate As, Cr, and Cd contents in rice because they posed greater non-carcinogenic and carcinogenic health risks than the others to the local residents.

Keywords Heavy metals · Rice · Spatial distribution · Health risk assessment

Introduction

With the rapid development of society and the improvement of people's living standards and quality, more and more social focus is on food security. China has been an agricultural power since ancient times and has a considerably long history of growing rice. More than half of the world's population is also dependent on rice as a staple food, most especially in China and its neighboring countries in South and Southeast Asia (Du et al. 2018; Ren et al. 2014). According to public statistics, the rice production in China

reached about 200 million tonnes in 2020, accounting for about 37% of the total global rice production and ranking the first in the world (Agriculture in China 2020). In particular, Hunan Province is known as “the hometown of fish and rice” with an annual rice production of 26.39 million tonnes, accounting for 12.45% of the national total (Agriculture in China 2020). Rice and its products are the main foodstuffs in this province, providing more than 70% of the total daily calorie intake for the local residents (Qian et al. 2010). However, Hunan Province is also known as the “hometown of non-ferrous metals” in terms of mineral species and mineral volume. The province has a proven number of 39 minerals, among which antimony (Sb) reserves are the first in the world (Long et al. 2019). The exhaust gas, wastewater, and slag from mining and smelting contribute to the increasing environmental risk (Wu et al. 2018). In recent years, there have been many reports about excessive heavy metal contents in rice from other cities and regions in China (Lu et al. 2021; Meng et al. 2014; Zhao et al. 2010), which has led to increasing public concern about food health safety. Therefore, the quality and safety of rice and local human health

Responsible Editor: Philippe Garrigues

✉ Jia Wen
jwen@hnu.edu.cn

¹ College of Environmental Science and Engineering, Hunan University, Changsha 410082, People's Republic of China

² Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha 410082, People's Republic of China

in Hunan as affected by the distinct regional characteristics should also be taken seriously.

Heavy metals are persistent, non-biodegradable, and potentially toxic to organisms (Kukusamude et al. 2021). Because of the long-term accumulation of heavy metals in Hunan Province, the metal concentrations in soil far exceed the local background value, which reduces the soil quality and causes significant changes in the soil ecosystem (Jia et al. 2020). Heavy metals in crops enter the human body through dietary intake, dermal contact, and respiration (Zheng et al. 2020). Compared to some other plants, rice is more likely to absorb heavy metals (Zhao et al. 2010). The uptake and accumulation of heavy metals by different tissues in rice differed in different growth periods, with the root system having the strongest sorption capacity and the accumulation order being grains < leaves < stems < roots (Chandra et al. 2018; Zhang et al. 2020a, b). For Hunan people, the absorption of heavy metals through the intake of contaminated rice is the main pathway that poses a potential risk to human health (Cai et al. 2015). The accumulation of heavy metals in the human body can lead to chronic heavy metal poisoning, causing metabolic disorders, respiratory diseases, and a series of mental illnesses and organ failures. These symptoms resulted from the stable biotoxicity of heavy metals through chelation with proteins or enzymes in the body (Fan et al. 2017; Lu et al. 2021).

Heavy metal contamination in polished rice grains in Hunan has been reported in a few studies by Chinese researchers. For example, Wang et al. (2017) studied the cationic heavy metal (i.e., cadmium (Cd), chromium (Cr), lead (Pb), zinc (Zn), nickel (Ni), copper (Cu), cobalt (Co)) accumulation in rice grains and analyzed their potential geological and health risks. The arable land in Hunan province is also known for severe anionic metal pollution, such as arsenic (As) and Sb (Liang et al. 2017; Okkenhaug et al. 2012). Especially, the world's largest Sb mine (Xi Kuang Shan) is located in Hunan Province, and Sb concentrations in the surrounding soil ranged from 100 to 5054 mg/kg (He 2007). However, studies that discussed about heavy metal concentrations in rice were only reported in some scattered mining areas in Hunan (Fan et al. 2017; Du et al. 2018). Besides, all these results were collected more than five years ago. Anionic heavy metal accumulation in rice grains in the wider area of Hunan has not yet been comprehensively investigated.

In this study, 34 rice grain samples were collected from local farmers and farmers' markets in 14 prefecture-level cities. Nine toxic heavy metals, including As, Cd, Cr, Pb, Zn, Ni, Cu, Co, and Sb, in the rice grain samples from Hunan Province, were analyzed. The objectives of this study were as follows: (1) to calculate the degree of heavy metal contamination in rice in Hunan Province based on the concentrations of the collected samples, (2)

to analyze the spatial distribution of heavy metals and their aggregation status, and (3) to perform the carcinogenic and non-carcinogenic risk assessment of the metals to human health. Data were subjected to principal component analysis and Pearson correlation analysis to determine their potential sources. The spatial distribution of the heavy metals was analyzed by ordinary kriging interpolation. This study will provide the latest information on the accumulation of heavy metals in rice grains and its spatial distribution in Hunan.

Materials and methods

Study area

The study area (108°47' – 114°15' E, 24°38' – 30°08' N) located in Hunan Province, China (Fig. 1). Hunan has a continental subtropical monsoonal humid climate with abundant water resources, with an average precipitation of 1450 mm over the years and a total water resource of 168.9 billion m³. The province is rich in land resources, with a total area of 211,800 km² and an arable land area of 4,148,800 ha (Hunan Provincial Government, 2020). The climate and topography provide perfect conditions for agricultural development, especially for rice cultivation. However, anthropogenic activities such as mineral extraction, smelting, urban development, and industrial expansion have caused heavy metal pollution problems (Wu et al. 2018). Therefore, rice quality and safety are of paramount importance to the permanent resident population of 69,183,800.

Sample collection and pretreatment

Thirty-four samples were collected from local farmers and farmers' markets in 14 prefecture-level cities of Hunan Province. Five sites were from Changsha and Huaihua, four from Yueyang, three from Yongzhou, Loudi, and Hengyang, two from Zhuzhou, Shaoyang, and Yiyang, and one from Changde, Xiangxi, Xiangtan, Chenzhou, and Zhangjiajie, respectively. The locations of sampling sites were recorded by using a global positioning system (GPS) locator (Table S1). At each sampling site, 2–3 samples were collected and mixed together, and one-quarter of the evened samples was taken out for metal analysis. All hulled rice samples were rinsed with deionized water to remove surface dust contaminants and then dried in an oven at 60 °C to a constant weight. Processed rice samples were crushed with a grinder and then passed through 100 mesh sieves (0.149 mm) and stored in airtight polyethylene bags until digestion.

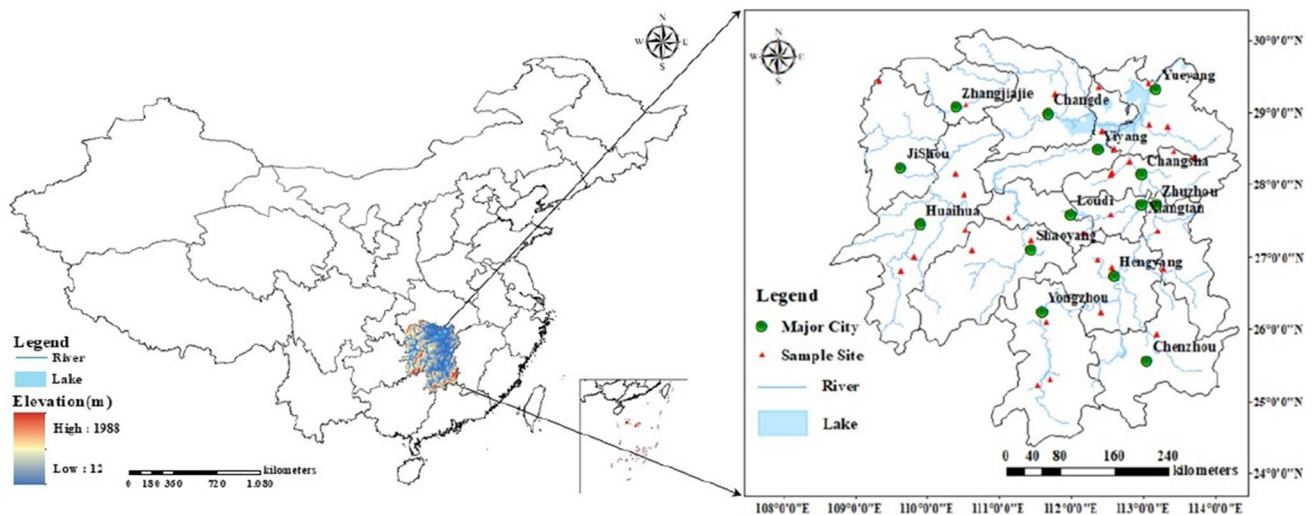


Fig 1. Study area and sampling sites.

Sample digestion and determination

Rice samples were digested using the graphite digestion method to identify the contained metals and contents (Du et al. 2018; Zhang et al. 2020a, b). All samples and the blank control were analyzed in triplicates. Before analysis, the dissolved solution was filtered through a 0.22 μm cellulose acetate membrane to remove any suspended matter (Ma et al. 2017). Nine target metals (i.e., As, Cd, Cr, Pb, Zn, Ni, Cu, Co, and Sb) were subsequently determined by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7900). The standard reference rice material used for analysis was Early indica rice LS/T 15121.1 (Guangzhou Testing Technology Co., LTD). Twenty-four elemental mixed standard solutions (100 $\mu\text{g}/\text{mL}$) used for calibration were purchased from the National Center for Analysis and Testing of Non-ferrous Metals and Electronic Materials (Beijing, China). Bismuth (Bi), indium (In), lutetium (Lu), rhodium (Rh), and terbium (Tb) were used as internal standards to make the calibration more accurate (Yuanan et al. 2020). The glassware and Teflon tubes used throughout the experiment were soaked in 5% HNO_3 solution for more than 24 h. They were repeatedly rinsed with deionized water and dried before use.

Data acquisition and analysis

In this study, the soil texture and population data were obtained from Institute of Geographic Sciences and Natural Resources Research, CAS Resource and Environment Science and Data Center. Soil property data (pH, soil organic carbon (SOC), cation exchange capacity (CEC)) were derived from National Earth System Science Data Center, China (Fig. S1). The measured data were used for statistically analyzed using SPSS 26.0, and contamination

assessment (including single-factor pollution index (SFPI) and Nemerov comprehensive pollution index (NCPI)) of the heavy metals were examined and analyzed (Supporting information Text 1). Classification of SFPI and NCPI of heavy metals is shown in Table S2 (Hu et al. 2017). The concentration data were also used to calculate non-carcinogenic risk and carcinogenic risk to humans (Supporting information Text 2). The potential sources of heavy metals in rice samples were analyzed by Pearson correlation analysis and principal component analysis (Ma et al. 2017). The spatial distribution of heavy metals in rice was analyzed by ordinary kriging interpolation in the ArcGIS 10.7 software. Figures were completed in the Origin 2018 and Excel 2019 software.

Results and discussions

Heavy metal concentrations in rice

The boxplot (Fig. 2a) showed the concentration of heavy metals in the rice grain. The dashed red line in the figure indicates the limit value (0.2 mg/kg) for Cd, Pb, and As; the dashed green line is the limit value (1.0 mg/kg) for Ni, Cr, and Sb; the dashed purple line is the limit value (10.0 mg/kg) for Cu; and the dashed blue line is the limit value (50.0 mg/kg) for Zn (NHC., 2017). The specific values and descriptive statistics are shown in Table 1. Among these metals, only the Cu, Zn, and Sb concentrations did not exceed their corresponding threshold value. There is no data regarding the limit concentration of Co in the Chinese standard; hence, it was not possible to evaluate the contamination status of Co. The mean values of Cr, As, Cd, and Pb were 1.28, 0.48, 0.28, and 0.66 mg/kg, respectively, which exceeded their individual limit concentration in rice (NHC., 2017). The

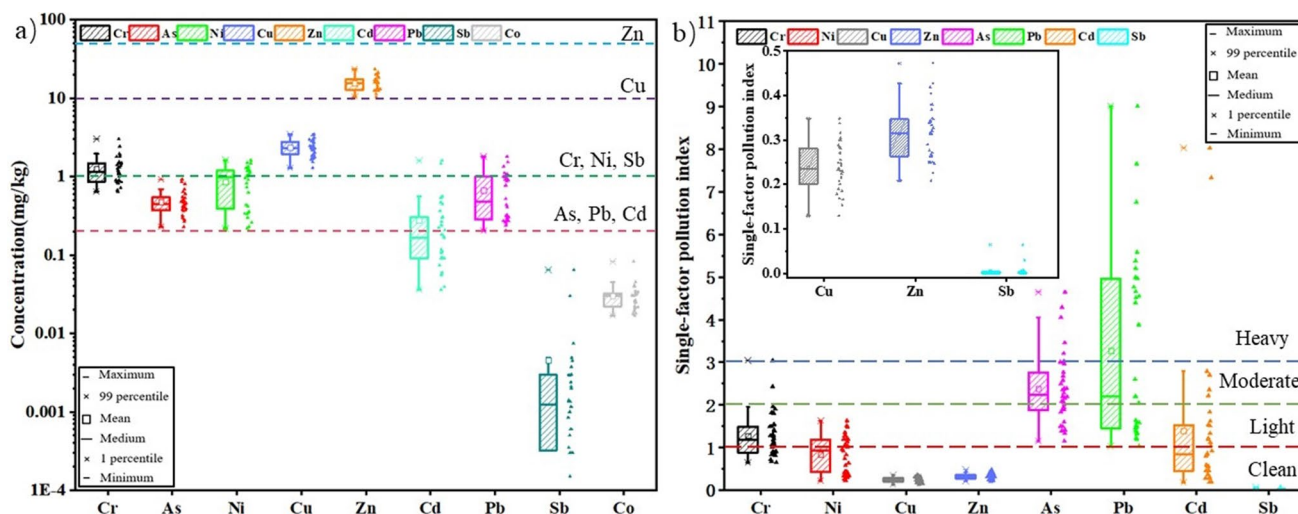


Fig 2. a Box plot of heavy metals concentration in rice; b box plot of single-factor pollution index of heavy metals in the rice grains

minimum concentrations of As (0.23) and Pb (0.21) were higher than their corresponding limit values, and thereby the exceedance rates were 100%. The maximum concentration of As (0.93) and Pb (1.80) in the rice grain was more than 4.0 times and 9.0 times higher than the limit value, respectively. The exceedance rates of Cr, Ni, and Cd were 64.70%, 47.05%, and 44.12%, respectively.

The normality of heavy metal concentrations in the rice grain was examined by the one-sample Kolmogorov-Smirnov (k-s) method (Zhou et al. 2014), and results were shown in Table 1. In this study, values of Cr, Co, Cu, Zn, As, Cd, and Sb all satisfied the normal distribution ($P > 0.05$), but Pb and Ni did not satisfy the requirements of normal distribution ($P \leq 0.05$), indicating that the concentration distributions of Pb and Ni at the sampling sites were significantly different (Wang et al. 2017). The asymmetry of the real-valued probability distribution was measured by skewness and kurtosis. The skewness of more than zero is positive skewness, with more values concentrating on the right side of the symmetry axis. Kurtosis describes the steepness of the distribution pattern, and the positive kurtosis values of Cr, Co, As, Cd, and Sb indicated a steeper than normal distribution (Yang et al. 2018). For some skewed data, logarithmic transformation is made to achieve a normal distribution (Sun et al. 2019). The coefficient of variation (CV) indicates the degree of dispersion and was used to evaluate the variation of heavy metal content and the level of differentiation in the sampling points. As shown in Table 1, the CVs for Cr, Co, Ni, Pb, and As were all above 30%, and the CVs of Cd and Sb were 126.3% and 276.5%, respectively, indicating high to very-high variation of the spatial distribution of these metals in the sampled rice grain. The factors responsible for the variation may be due to various anthropogenic activities in each sampling region (Li et al. 2012; Ma et al. 2017). At the sampling sites, the nature of the soil itself (Fig. S1) is quite

different in terms of textures and properties, which affects the migration of heavy metals in the soil and the amount of heavy metals taken up by the rice.

Sb and As are from the same group (VA) in the periodic table; thus, the two show many chemical similarities. Fan et al. (2017) found that both Sb (5.175 mg/kg) and As contents (0.524 mg/kg) in rice near a mine area were higher than the Sb (0.004 mg/kg) and As (0.48 mg/kg) contents in rice in this study. It indicates that there is basically no threat of Sb pollution in the greater region of Hunan except for mining areas, but As pollution are above the limit value and it is a hazard that quick reaction should be taken. For cationic metals, the concentrations of Ni, Cu, Zn, Co, and Cd were similar to those of related studies. Ni values were relatively high with parts above the limit value and Cd was all above the limit value, and Cu, Zn, and Co were within the limit values. We found a similar Cd concentration in Changsha rice (0.283 mg/kg) to that in the study of Du et al. (2018) (0.291 mg/kg), suggesting that the Changsha is a central pollution area affected by Cd, and the possible cause is the highly concentrated manufacturing. In addition, the Cr and Pb concentrations in this study were higher than those in the relevant studies (Du et al. 2018; Wang et al. 2017). The uptake of Cd, Cr, and Pb in rice was also proportional to the increase in soil heavy metal content (Wang et al. 2016), thus bringing new alerts to the local regulators and producers to take action on soil remediation and food safety monitoring.

Contamination assessment of heavy metals in rice grain

The pollution level of heavy metals was assessed according to Table S2. Among them, the average single factor pollution index (SFPI) of heavy metals followed the order of

Table 1 Descriptive statistics of heavy metal concentrations in rice grains from Hunan Province.

Element	Max (mg/kg)	Min (mg/kg)	Median (mg/kg)	SD	Mean (mg/kg)	CV	Over-standard rate	K-S (normal)p	Skewness	Turtosis	Threshold value (mg/kg)
Cr	3.04	0.64	1.21	0.52	1.28	40.7%	64.7%	0.188	1.50	2.97	1.00 ^b
Co	0.083	0.011	0.030	0.012	0.03	40.9%	/	0.121	2.62	11.17	Null ^a
Ni	1.63	0.22	0.93	0.43	0.84	51.6%	47.1%	0.004	0.02	-1.39	1.00 ^b
Cu	3.49	1.29	2.35	0.53	2.39	22.3%	0%	0.200	0.06	-0.47	10.00 ^b
Zn	23.64	10.44	15.71	3.13	15.73	19.9%	0%	0.200	0.53	-0.12	50.00 ^b
As	0.93	0.23	0.46	0.17	0.48	35.0%	100%	0.200	1.01	0.95	0.20 ^b
Cd	1.61	0.04	0.17	0.35	0.28	126.3%	44.1%	0.200	2.95	9.09	0.20 ^b
Pb	1.80	0.21	0.44	0.43	0.66	65.8%	100%	0.001	0.85	-0.11	0.20 ^b
Sb	0.065	0.00	0.001	0.012	0.004	276.5%	0%	0.200	4.57	22.01	1.00 ^c

^a“Null,” no relevant standard value was found.^bThe values from National Standards for Food Safety (NHC,2017).^cPublic Health and Municipal Services Ordinance of Hong Kong (PHMSO).

Pb (3.279) > As (2.384) > Cd (1.389) > Cr (1.278) > Ni (0.839) > Zn (0.315) > Cu (0.239) > Sb (0.004). According to the standards, there was likely pollution when the SFPI was more than 1.0 (Table S2). As shown in Fig. 2b, the average SFPI of Pb, As, Cd, and Cr was more than 1.0. The proportion of SFPI pollution levels of each heavy metal in all sampling sites was shown in Fig. 3a, with the exceedance rates of Pb, As, Cd, and Cr being 100%, 100%, 44.12%, and 64.71%, respectively. Moderate pollution and heavy pollution sites accounted for 52.94%, 67.65%, 17.64%, and 5.88%, respectively. Therefore, it can be concluded that the main sources of metal contamination in rice in Hunan Province were Pb, As, Cd, and Cr (Zhang et al. 2018a, b). For Ni, 52.94% of the sampling sites were lightly polluted, and the rest were not contaminated. For Cu, Zn, and Sb, the SFPI were all below 1.0; hence, the rice grain collected from the 34 sampling sites was safe for these three metals. We also compared the sampling values with Chinese standards (NHC., 2017; PHMSO) and generated the NCPI of all metals in the 34 sampling sites (Fig. 3b). Sixteen rice samples (47.06%) were lightly polluted with $1 < \text{NCPI} \leq 2$. Three rice samples (8.82%) were moderately polluted with $2 < \text{NCPI} \leq 3$. Fifteen rice samples (44.12%) were heavily polluted with $\text{NCPI} \geq 3$. The spatial distribution of the NCPI values is shown in Fig. 3c. It can be seen from the legend that NCPI values were all over 1.0, and the pollution degree is light pollution or above. Severely contaminated areas were mainly found in the central and northeastern parts of Hunan Province, such as Shaoyang and Changsha.

Spatial distribution of heavy metals in rice

The spatial distribution of heavy metals in rice samples was analyzed by the ordinary kriging interpolation method in the GIS software. As shown in Fig. 4a and b, the spatial distribution of As and Pb was relatively similar, with higher concentrations in the central-eastern and southwestern parts of Hunan Province and were relatively lower in other areas. The heaviest pollution of As and Pb in Changsha, Zhuzhou, and Xiangtan urban agglomerations might be due to their industrial production (Ma et al. 2016). In addition, the large amount of wastewater and waste gas generated by local industries, mining, etc., has greatly increased the amount of heavy metals in the environment, thus leading to rice pollution (Xiao et al. 2018).

As shown in Fig. 4c, d, and e, Cr, Zn, and Cu also showed similar spatial distribution patterns, with higher concentrations mainly concentrated in the central and northern regions, such as Hengyang, Shaoyang, and Changsha. It was similar to the results reported by Wang et al. (2017). Since there are few studies on the spatial distribution of metals in rice in Hunan Province, this study has certain positive significance. The concentration ranges of Zn (10.44–23.64 mg/

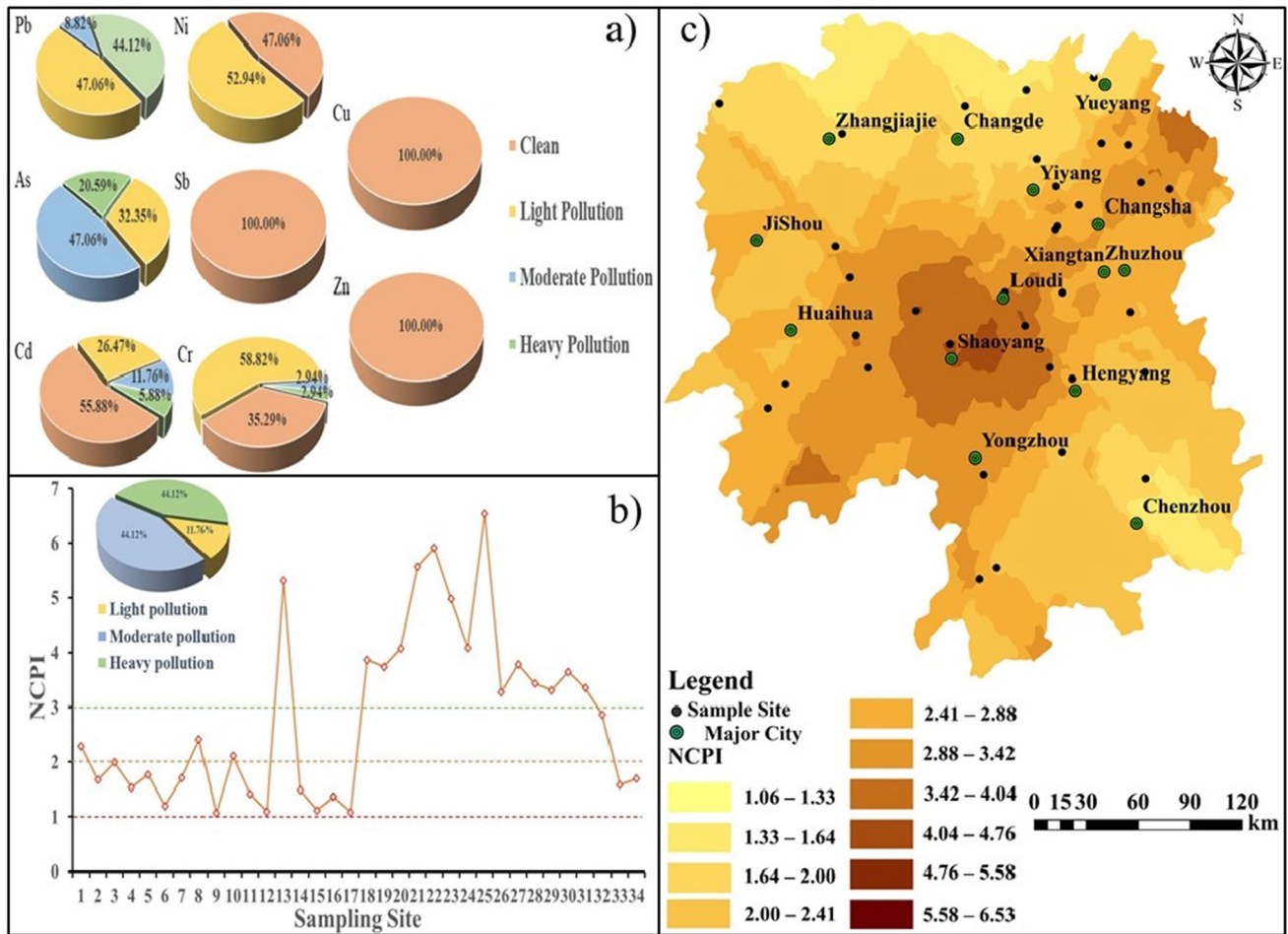


Fig 3. The proportion of pollution levels of each heavy metal element in all sampling sites: **a** proportion of SFPI pollution levels; **b** proportion of NCPI pollution values and levels. The red, orange, and green

dashed lines represent NCPI values of 1.0, 2.0, and 3.0, respectively; and **c** spatial distribution of NCPI in Hunan Province.

kg) and Cu (1.29–3.49 mg/kg) were below their respective standard limits (50.0 mg/kg for Zn and 10.0 mg/kg for Cu), and therefore, Cu and Zn were relatively safe for humans. However, 64.7% of Cr in the samples exceeded the limit value (1.0 mg/kg) (Table 1), indicating that Cr pollution was widespread in Hunan Province.

The spatial distribution of Cd is in the central and southern regions (Fig. 4f), with 44.1% of rice samples exceeding the Cd concentration limit (0.2 mg/kg). The results of the study were consistent with the result of Xiao et al. (2018), indicating that Cd pollution has been widespread in Hunan Province. Relatively high concentrations of Co were in the southern region (Fig. 4g), and the higher concentrations of Ni were in the central and western areas (Fig. 4h), such as Loudi. It is basically consistent with the result of Wang et al. (2017). Interestingly, the Ni (0.84 mg/kg) concentration in Loudi was lower than that of Zijiang River basin rice (1.33 mg/kg) (Zhang et al. 2020a, b) but was higher than that in Changsha (0.343 mg/kg) (Du et al. 2018), possibly due to

sewage irrigation in Zijiang river basin and Loudi region. The relatively high concentrations of Sb were in the central and northwestern parts of the region (Fig. 4i), and all samples were below the limit values (1.0 mg/kg). The reason might be that Sb mining in Hunan Province is mainly concentrated in Loudi, but the sampling sites were not around the mining areas, so the impact was relatively low. Relating to the results of spatial distribution of heavy metals, it was also found that the areas with a higher concentration of heavy metals had denser population distribution (Fig. S2).

Potential sources of heavy metals in the rice samples

Pearson correlation coefficient method and principal component analysis were used to determine the correlation and potential sources of heavy metals (Chai et al. 2017; Zhang et al. 2018a, b). Highly significant positive correlations ($P \leq 0.01$) were found in Cr-Cu-Zn, As-Co-Zn, and

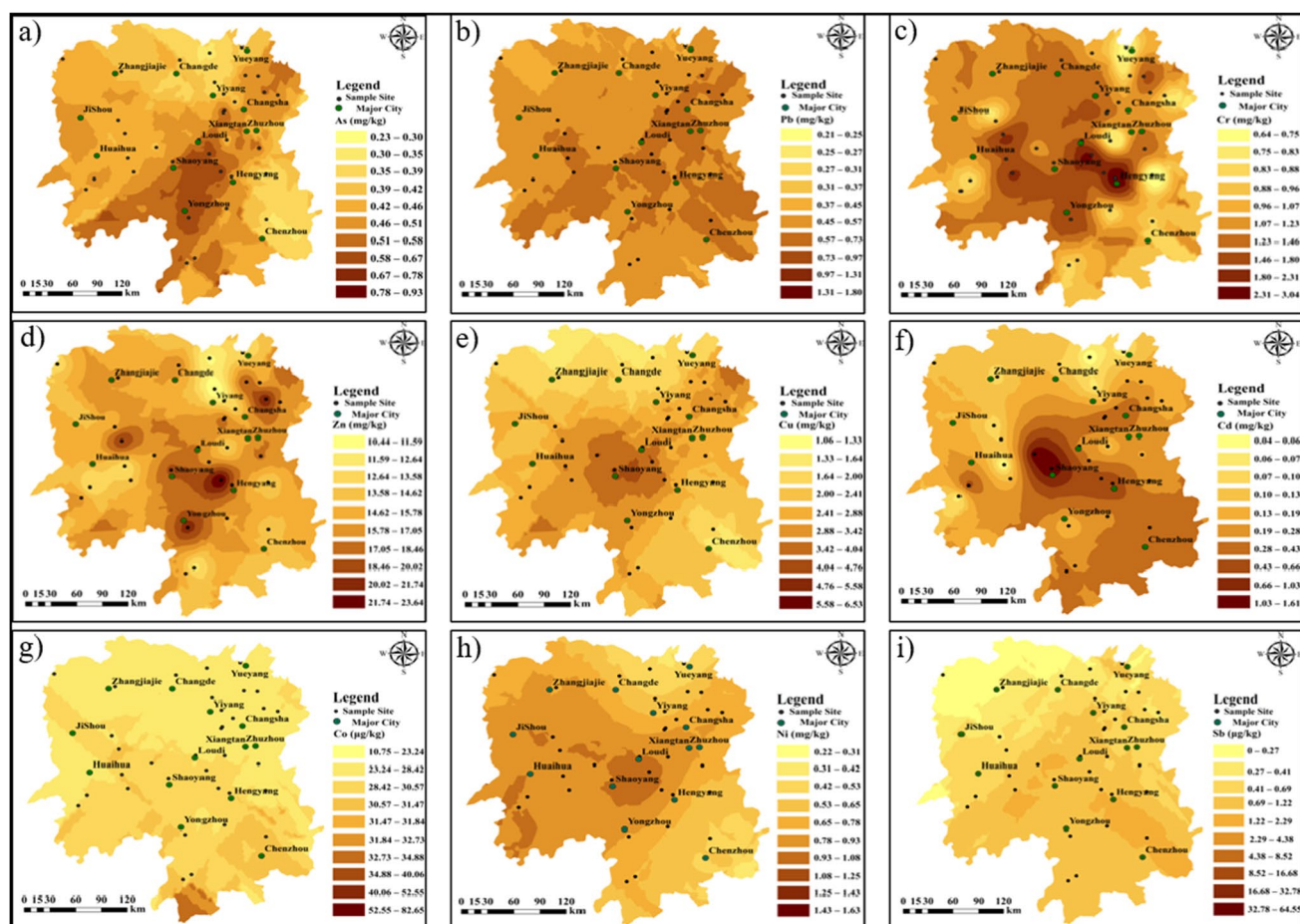


Fig. 4. Spatial distribution map of heavy metals concentration in rice grains from Hunan Province

Ni-Zn-As-Pb combinations (Fig. 5a and Table S3). Significant positive correlations ($P \leq 0.05$) were found in the Cr-As-Co, Co-Ni-Cu, and Cu-As combinations. The strength of the correlations indicates the existence of a common source among the heavy metals to some extent. For example, Pb and As can originate from the natural environment and the chemicals used in agriculture (e.g., compound fertilizers and Pb/As pesticides). The use of chemical fertilizers and pesticides in southern China has a history of high usage. According to the statistics, the number of chemical fertilizers and pesticides applied in Hunan Province in the single year of 2020 reached 7,330,120 tonnes and 101,450 tonnes, respectively (Hunan Statistical Yearbook., 2021). Therefore, heavy metal pollution of soil in Hunan due to agricultural activities is severe. Besides, Co and Cu come from natural sources and forage, and they enter the soil with organic fertilizer; Pb, Zn, and Ni can also be derived from fossil fuel combustion and mining, such as Pb-Zn ores found in many places in Hunan (China Geological Survey., 2021).

Using principal component analysis (PCA) further determines the source of heavy metals in rice. The KMO values (0.737 > 0.5) and Bartlett's test (0.000 < 0.001) proved the validity of the PCA method (Wang et al. 2017). The proportion of metal components and eigenvalues were presented in Table S4 and Fig. 5b, explaining 63.1% of the total variance. The first principal component (PC1) accounted for 48.5% of the total variance, in which Cr, Co, Ni, Cu, Zn, As, and Pb were the main contributors. The results were consistent with Pearson correlation coefficients, where these metals were closely correlated with each other. The inclusion of multiple metals in the PC1 may be derived from diverse activities that concurrently produced them, such as agricultural production, industrial production, and mining. The second principal component (PC2) accounted for 14.6% of the total variance, in which Cd and Sb were included. Fertilizer and pesticide abuse are possibly the main sources of Cd contamination (Wang et al. 2015). Sb mainly originates from the mining and smelting of minerals, such as the Xikuangshan in Loudi and Yuanling County, Huaihua. Sb can be easily absorbed by plant roots, and the rice around Xikuangshan smelter was

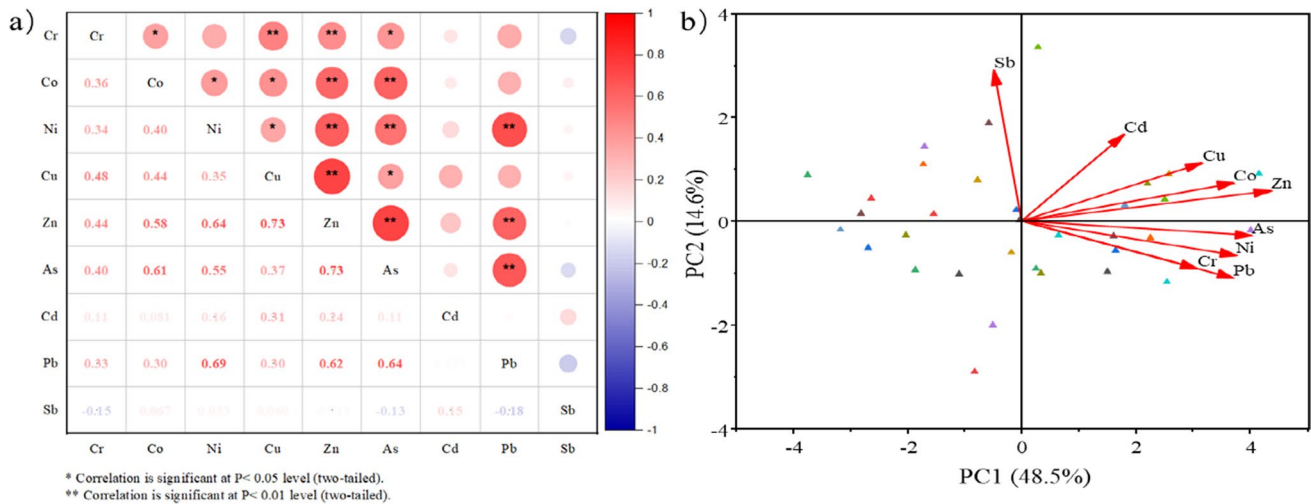


Fig 5. a Pearson’s correlation matrix for heavy metal concentrations; b principal component analysis of heavy metals in rice of Hunan Province

reported to accumulate high concentrations of Sb up to 225 mg/kg in the roots and 5.79 mg/kg in the seeds, suggesting serious pollution (Ren et al. 2014). Nevertheless, our study did not find widespread Sb contamination in the rice grain, implying Sb contamination only occurred as point-source pollution.

Health risks of heavy metals in rice

Estimated daily intake of heavy metals via rice

The average estimated daily intake of heavy metals for adults and children in Hunan is shown in Table 2. Although adults consume high amounts of rice, most studies have also reported higher metal intakes in children than in adults due to their low body weight (Zhang et al. 2020a, b; Lu et al. 2021). In this study, the estimated daily intake (EDI) for

children was 1.27 times higher than that for adults. In terms of the total amount, the heavy metal intake of Hunan people followed the sequence of Zn > Cu > Cr > Ni > Pb > As > Cd > Co > Sb. Although Zn and Cu were ranked as the top two metals for human intake, their nutritious role in the metabolic system relieves the concerns on the intake quantity, and the vast area of Hunan Province was not found with Cu and Zn contamination in rice (Fig. 3a). In contrast, Cr and Ni intake should be paid more attention because these two metals were found with moderate contamination in the rice grain.

Non-carcinogenic risks

In this study, the assessment of potential health risk of different heavy metals in the rice grain was determined by target hazard quotient (THQ) and hazard index (HI), where

Table 2 Assessment results of health risk in rice grains (mean values are given).

Elements	RfD(mg·kg ⁻¹ ·d ⁻¹)	SF(kg·d·kg ⁻¹)	EDI(mg·kg ⁻¹ ·d ⁻¹)		THQ		CR	
			Children	Adults	Children	Adults	Children	Adults
Cr	3.000×10 ⁻³	0.5×10 ⁻²	1.188×10 ⁻²	9.346×10 ⁻³	3.962	3.115	5.942×10 ⁻³	4.673×10 ⁻³
Co	3.000×10 ⁻⁴	/	2.739×10 ⁻⁴	2.154×10 ⁻⁴	0.913	0.718	/	/
Ni	2.000×10 ⁻²	1.7	7.808×10 ⁻³	6.140×10 ⁻³	0.390	0.307	1.327×10 ⁻²	1.044×10 ⁻²
Cu	4.000×10 ⁻²	/	2.225×10 ⁻²	1.753×10 ⁻²	0.556	0.438	/	/
Zn	3.000×10 ⁻¹	/	1.463×10 ⁻¹	1.150×10 ⁻¹	0.488	0.384	/	/
As	3.000×10 ⁻⁴	1.5	4.435×10 ⁻³	3.488×10 ⁻³	14.783	11.626	6.652×10 ⁻³	5.232×10 ⁻³
Cd	1.000×10 ⁻³	6.1	2.581×10 ⁻³	2.030×10 ⁻³	2.581	2.030	1.575×10 ⁻²	1.238×10 ⁻²
Pb	3.500×10 ⁻³	8.500×10 ⁻³	6.100×10 ⁻³	4.797×10 ⁻³	1.743	1.371	5.185×10 ⁻⁵	4.077×10 ⁻⁵
Sb	4.000×10 ⁻³	/	3.981×10 ⁻⁵	3.130×10 ⁻⁵	0.010	0.008	/	/

SF, carcinogenic slope factors [(kg·d)/mg].

CR, carcinogenic risk.

THQ and $HI \geq 1.0$ are associated with higher risk to human health (Ma et al. 2016). As shown in Table 2, the calculated mean values of THQ for adults and children were $As > Cr > Cd > Pb > 1.0 > Co > Cu > Zn > Ni > Sb$. As has the largest THQ value, which is similar to the results found in the Zijiang basin of Hunan Province (Zhang et al. 2020a, b) and Longyan of Fujian Province (Lu et al. 2021). In addition, the minimum values of As and Cr were higher than 1.0 (Table S5), indicating that prolonged intake of rice As and Cr has posed a serious potential non-carcinogenic risk to the health of Hunan residents (Ma et al. 2017). The mean values of THQ for Cd and Pb were all higher than 1.0, while the minimum values were all less than 1.0 (Table S5). The results of THQ_{Cd} value in the present study (2.131) were similar to the results in Du et al. (2017) (2.030), indicating considerable potential non-carcinogenic health risk of Cd to local inhabitants from rice consumption. The mean and maximum values of THQ for Co, Cu, Zn, Ni, and Sb were all less than 1.0, implying that there was a little non-carcinogenic risk to human health from these metals in Hunan Province. In particular, both Cu and Zn are essential minerals for human survival, but excessive intake can cause adverse health effects (Vatanpour et al. 2020).

Due to the accumulation of heavy metals in crops, there is often an accumulation of multiple metals in organisms through the food chain. As shown in Fig. 6a, the HI of heavy

metals for adults and children was greater than 1.0 in all prefecture-level cities and exceeded the limit nearly 10.9–26.0 and 13.8–33.1 times, respectively, implying that the Hunan residents' health may be at risk due to the consumption of polished rice (Lei et al. 2015). In particular, the relative contributions of As (58.14%), Cr (15.58%), and Cd (10.15%) were greater than the others (Fig. 6b). The relatively high non-carcinogenic risks were concentrated in Shaoyang, Loudi, Hengyang, Changsha, and Yongzhou, indicating that residents in these areas may suffer much higher risks of heavy metal-induced diseases.

Carcinogenic risks

Carcinogenic risk (CR) refers to the higher-than-normal level of cancer incidence due to lifetime exposure to carcinogens. The acceptable range of CR is from 10^{-6} to 10^{-4} (USEPA 2006). In this study, only five metals (i.e., Cr, Ni, As, Pb, and Cd) were calculated for the CR values because slope factors (SF) of Co, Cu, Zn, and Sb were not provided in any published database or literature. The CR values of Cr, Ni, As, and Cd were all higher than the acceptable range for both adults and children in this study (Table 2), with CR in the order of $Cd > Ni > As > Cr$. However, the values in this study were greater than that in Zeng et al. (2015), indicating a growing potential carcinogenic risk of Cd, Ni,

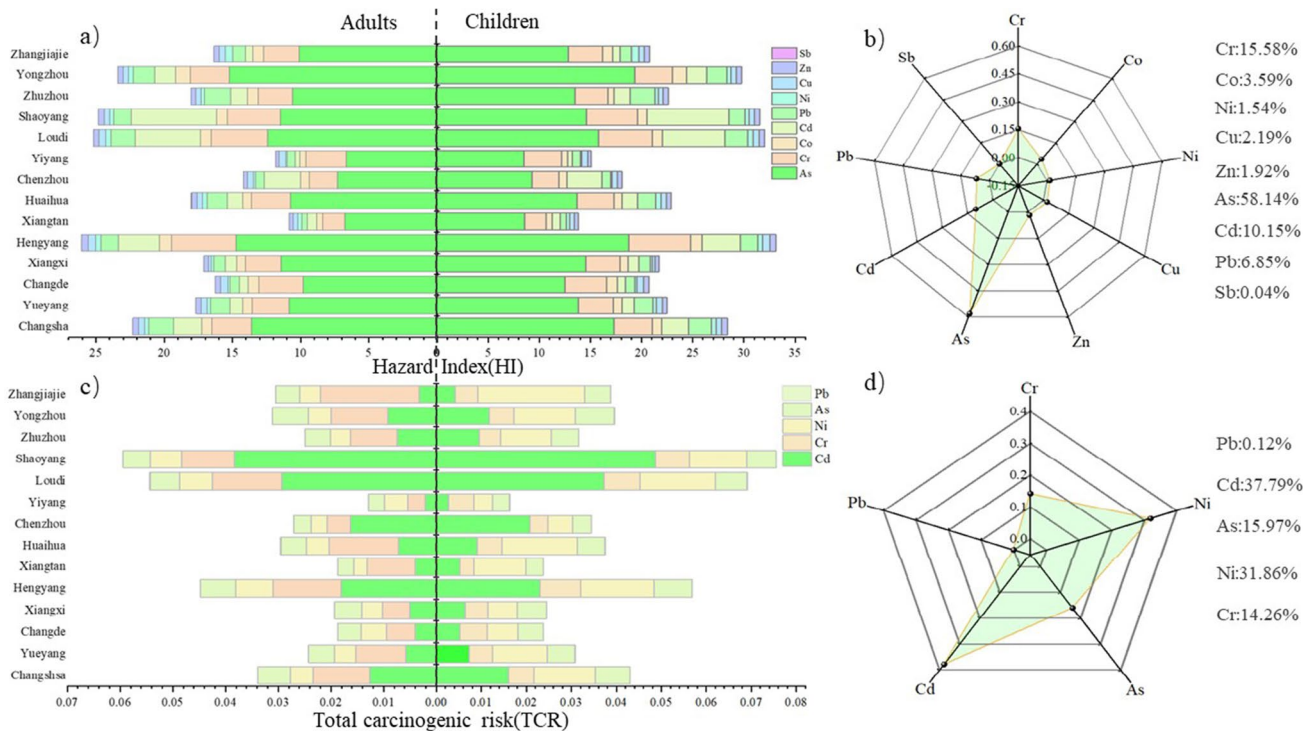


Fig. 6. a Heavy metal hazard index and b the percentage of hazard heavy metal contribution in rice grains; c total carcinogenic risk values of heavy metals; and d the percentage carcinogenic risk contribution of heavy metals in rice grains

As, and Cr through rice consumption in Hunan Province with time. Only Pb was within the acceptable range. The total carcinogenic risk (TCR) of the five heavy metals in rice ranged from 0.0128–0.0595 for adults and 0.0163–0.0756 for children, respectively (Fig. 6c). The carcinogenic risk of Cd was higher than that of Ni and As, which is the same as that of Zhang et al. (2020a, b). Besides, Cd, Ni, As, and Cr were the major carcinogenic metals that accounted for 37.79%, 31.86%, 15.97%, and 14.26% of the TCR, respectively (Fig. 6d). Therefore, more efforts should be paid to lower down Cd, Ni, As, and Cr concentrations in the rice and soil through various technologies and production management. The top four prefecture-level cities in terms of TCR were Shaoyang (adults: 0.0596, children: 0.0756) > Loudi (adults: 0.0544, children: 0.0692) > Hengyang (adults: 0.0448, children: 0.0569) > Changsha (adults: 0.0339, children: 0.0431) (Fig. 6c), requiring priority in the regulation of pollution control and food supervision.

In conclusion, the health risks associated with heavy metals in children were higher than in adults. Besides dietary intake, children's specific habits (pica behavior, crawling on the ground, and hand or finger sucking) can also increase the risk (Tan et al. 2016). The health risk assessment results showed that long-term consumption of local rice in Hunan Province has a significant carcinogenic risk to human health, such as increasing the risk of lung cancer and mental illnesses; the incidence of lung cancer in Hunan province was 55.60/100,000, ranking first among all cancers (Li et al. 2021). Studies have shown that Cd and Pb could increase the incidence of gastric cancer and colon cancer, respectively, and the incidence will be higher when multiple metals are combined (Fei et al. 2018; Kiani et al. 2021; Zhao et al. 2014).

Conclusions

The analysis of 9 heavy metals (i.e., Cr, Ni, As, Pb, Cd, Co, Cu, Zn, and Sb) in the rice grain from Hunan Province showed the cumulative and hidden nature of rice contamination in the region. Among these metals, Pb and As accumulation in rice were the most serious. According to Pearson correlation analysis and principal component analysis, potential pollution sources were identified: Cr, As, Sb, and Pb originated from mineral extraction, smelting, and industrial activities; Co, Ni, Cd, Zn, and Cu originated from mining, agricultural activities, and forage. Health risk assessment showed that hazard index and total carcinogenic risk were much higher than the acceptable range. In particular, the HI of As, Cr, Cd, and Pb posed a serious potential non-carcinogenic risk to the health of Hunan residents; the CR of Cd, Ni, As, and Cr accounted for relatively large proportions in the TCR and showed a growing trend as compared

to previous results. The study may provide the government and related agencies with guidance on regulating food safety. For example, residents should be advised to diversify their food selection so as to reduce their daily consumption of local rice; care should be taken not only on the rice but also the rice-made products. In addition, urgent reaction should be taken to reduce the sources of contamination and prevent and control the assimilation of heavy metals through food chain.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s11356-022-21636-6>.

Acknowledgements Acknowledgement for Dr. Yejia Lin from Geological Survey of Hunan Institute for GIS data acquisition. The data support from the National Earth System Science Data Center, National Science & Technology Infrastructure of China. (<http://www.geodata.cn>).

Data availability The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Author contribution All authors contributed to the study conception and design: Hongsheng Cui: methodology, investigation, experiment, data curation, writing—original draft; Jia Wen: conceptualization, funding acquisition, methodology, supervision, project administration; Lisha Yang: experiment, writing—review and editing; Qi Wang: experiment, data curation; meanwhile, all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

References

- Agricultural in china, 2020. Available at: <http://www.agdata.cn/newsdata/getagdatanewsdetails-733.html>
- Cai LM, Xu ZC, Qi JY, Feng ZZ, Xiang TS (2015) Assessment of exposure to heavy metals and health risks among residents near Tonglushan mine in Hubei, China. *Chemosphere* 127:127–135. <https://doi.org/10.1016/j.chemosphere.2015.01.027>
- Chai L, Li H, Yang Z, Min X, Liao Q, Liu Y, Men S, Yan Y, Xu J (2017) Heavy metals and metalloids in the surface sediments of the Xiangjiang River, Hunan, China: distribution, contamination, and ecological risk assessment. *Environ Sci Pollut Res Int* 24(1):874–885. <https://doi.org/10.1007/s11356-016-7872-x>
- China Geol Survey, 2021. Available at: <https://geocloud.cgs.gov.cn/>
- Chandra R, Kumar V, Tripathi S, Sharma P (2018) Heavy metal phyto-extraction potential of native weeds and grasses from endocrine-disrupting chemicals rich complex distillery sludge and their histological observations during in-situ phytoremediation. *Ecol Eng* 111:143–156. <https://doi.org/10.1016/j.ecoleng.2017.12.007>

- Dai W, Zhao K, Fu W, Jiang P, Li Y, Zhang C, Gielen G, Gong X, Li Y, Wang H, Wu J (2018) Spatial variation of organic carbon density in topsoils of a typical subtropical forest, southeastern China. *Catena* 167:181–189. <https://doi.org/10.1016/j.catena.2018.04.040>
- Du F, Yang Z, Liu P, Wang L (2018) Accumulation, translocation, and assessment of heavy metals in the soil-rice systems near a mine-impacted region. *Environ Sci Pollut Res Int* 25(32):32221–32230. <https://doi.org/10.1007/s11356-018-3184-7>
- Fan Y, Zhu T, Li M, He J, Huang R (2017) Heavy metal contamination in soil and brown rice and human health risk assessment near three mining areas in central China. *J Healthc Eng* 2017:4124302. <https://doi.org/10.1155/2017/4124302>
- Fei X, Lou Z, Christakos G, Ren Z, Liu Q, Lv X (2018) The association between heavy metal soil pollution and stomach cancer: a case study in Hangzhou City, China. *Environ Geochem Health* 40(6):2481–2490. <https://doi.org/10.1007/s10653-018-0113-0>
- Fu Z, Wu F, Mo C, Deng Q, Meng W, Giesy JP (2016) Comparison of arsenic and antimony biogeochemical behavior in water, soil and tailings from Xikuangshan, China. *Sci Total Environ* 539:97–104. <https://doi.org/10.1016/j.scitotenv.2015.08.146>
- Gao Z, Fu W, Zhang M, Zhao K, Tunney H, Guan Y (2016) Potentially hazardous metals contamination in soil-rice system and its spatial variation in Shengzhou City, China. *J Geochem Explor* 167:62–69. <https://doi.org/10.1016/j.gexplo.2016.05.006>
- Hunan Provincial Government. 2020. Hunan province in China., Available at: <http://www.hunan.gov.cn/hnszf/jxxx/hngk/zrdl/zrdl.html>
- Hunan Statistical Yearbook., 2021. Hunan Provincial Bureau of Statistics. Available at: <http://222.240.193.190/2021tjnj/indexch.htm>
- He M (2007) Distribution and phytoavailability of antimony at an antimony mining and smelting area, Hunan, China. *Environ Geochem Health* 29(3):209–219. <https://doi.org/10.1007/s10653-006-9066-9>
- He M, Wang X, Wu F, Fu Z (2012) Antimony pollution in China. *Sci Total Environ* 421–422:41–50. <https://doi.org/10.1016/j.scitotenv.2011.06.009>
- Hu B, Jia X, Hu J, Xu D, Xia F, Li Y (2017) Assessment of heavy metal pollution and health risks in the soil-plant-human system in the Yangtze River Delta, China. *Int J Environ Res Public Health* 14(9):1042. <https://doi.org/10.3390/ijerph14091042>
- Jia X, Fu T, Hu B, Shi Z, Zhou L, Zhu Y (2020) Identification of the potential risk areas for soil heavy metal pollution based on the source-sink theory. *J Hazard Mater* 393:122424. <https://doi.org/10.1016/j.jhazmat.2020.122424>
- Kiani B, Hashemi Amin F, Bagheri N, Bergquist R, Mohammadi AA (2021) Yousefi, Association between heavy metals and colon cancer: an ecological study based on geographical information systems in North-Eastern Iran. *BMC Cancer* 21(1):414. <https://doi.org/10.1186/s12885-021-08148-1>
- Kukusamude C, Sricharoen P, Limchoowong N, Kongsri S (2021) Heavy metals and probabilistic risk assessment via rice consumption in Thailand. *Food Chem* 334:127402. <https://doi.org/10.1016/j.foodchem.2020.127402>
- Lei M, Tie B-q, Song Z-g, Liao B-H, Lepo JE, Huang Y-z (2015) Heavy metal pollution and potential health risk assessment of white rice around mine areas in Hunan Province, China. *Food Sec* 7(1):45–54. <https://doi.org/10.1007/s12571-014-0414-9>
- Li C, Sun G, Wu Z, Zhong H, Wang R, Liu X, Guo Z, Cheng J (2019) Soil physiochemical properties and landscape patterns control trace metal contamination at the urban-rural interface in southern China. *Environ Pollut* 250:537–545. <https://doi.org/10.1016/j.envpol.2019.04.065>
- Li C, Wang J, Liao X-z, Xu K-k, Zou Y-h, Xiao H-f, Shi Z-h, Cao S-y, Guo J, Wang S-y, Li N, Yan S-p (2021) Trend of incidence and mortality of lung cancer in Hunan cancer registration areas from 2013 to 2017. *China. Cancer* 30(11):841–847. <https://doi.org/10.11735/j.issn.1004-0242.2021.11.A007>
- Li Q, Chen Y, Fu H, Cui Z, Shi L, Wang L, Liu Z (2012) Health risk of heavy metals in food crops grown on reclaimed tidal flat soil in the Pearl River Estuary, China. *J Hazard Mater* 227–228:148–154. <https://doi.org/10.1016/j.jhazmat.2012.05.023>
- Liang J, Feng C, Zeng G, Gao X, Zhong M, Li X, Li X, He X, Fang Y (2017) Spatial distribution and source identification of heavy metals in surface soils in a typical coal mine city, Lianyuan, China. *Environ Pollut* 225:681–690. <https://doi.org/10.1016/j.envpol.2017.03.057>
- Long J, Tan D, Deng S, Li B, Ding D, Lei M (2019) Antimony accumulation and iron plaque formation at different growth stages of rice (*Oryza sativa* L.). *Environ Pollut* 249:414–422. <https://doi.org/10.1016/j.envpol.2019.03.042>
- Lu Q, Xiao Q, Wang Y, Wen H, Han B, Zheng X, Lin R (2021) Risk assessment and hotspots identification of heavy metals in rice: a case study in Longyan of Fujian province, China. *Chemosphere* 270:128626. <https://doi.org/10.1016/j.chemosphere.2020.128626>
- Ma L, Wang L, Jia Y, Yang Z (2016) Arsenic speciation in locally grown rice grains from Hunan Province, China: spatial distribution and potential health risk. *Sci Total Environ* 557–558:438–444. <https://doi.org/10.1016/j.scitotenv.2016.03.051>
- Ma L, Wang L, Tang J, Yang Z (2017) Arsenic speciation and heavy metal distribution in polished rice grown in Guangdong Province, Southern China. *Food Chem* 233:110–116. <https://doi.org/10.1016/j.foodchem.2017.04.097>
- Meng B, Feng X, Qiu G, Anderson CW, Wang J, Zhao L (2014) Localization and speciation of mercury in brown rice with implications for pan-Asian public health. *Environ Sci Technol* 48(14):7974–7981. <https://doi.org/10.1021/es502000d>
- Mao C, Song Y, Chen L, Ji J, Li J, Yuan X, Yang Z, Ayoko GA, Frost RL, Theiss F (2019) Human health risks of heavy metals in paddy rice based on transfer characteristics of heavy metals from soil to rice. *Catena* 175:339–348. <https://doi.org/10.1016/j.catena.2018.12.029>
- Montserrat F, Nelson B, Yu W (2002) Antimony in the environment: a review focused on natural waters I. Occurrence. *Earth-Sci Rev* 57:125–176
- N. T. Basta, J.A.R., and R. L. Chaney, 2005. Trace element chemistry in residual-treated soil: key concepts and metal bioavailability. *J Environ Qual* 34(1), 49–63. DOI <https://doi.org/10.2134/jeq2005.0049dup>
- National Earth System Science Data Center, n.d. National Science & Technology Infrastructure of China. Available at: (<http://www.geodata.cn>)
- NHC, 2017. National Food Safety Standards Limits of Contaminants in Food (GB2762—2017). National Health Commission of the PRC, Beijing, China. Available at: <https://sppt.cfsa.net.cn:8086/db>
- Okkenhaug G, Zhu YG, He J, Li X, Luo L, Mulder J (2012) Antimony (Sb) and arsenic (As) in Sb mining impacted paddy soil from Xikuangshan, China: differences in mechanisms controlling soil sequestration and uptake in rice. *Environ Sci Technol* 46:3155–3162. <https://doi.org/10.1021/es2022472>
- Pierart A, Shahid M, Sejalon-Delmas N, Dumat C (2015) Antimony bioavailability: knowledge and research perspectives for sustainable agricultures. *J Hazard Mater* 289:219–234. <https://doi.org/10.1016/j.jhazmat.2015.02.011>
- PHMSO. n.d. Public Health and Municipal Services Ordinance of Hong Kong. Available at: <https://www.elegislation.gov.hk/hk/cap132/en>
- Qian Y, Chen C, Zhang Q, Li Y, Chen Z, Li M (2010) Concentrations of cadmium, lead, mercury and arsenic in Chinese market milled rice and associated population health risk. *Food Control* 21(12):1757–1763. <https://doi.org/10.1016/j.foodcont.2010.08.005>
- Ren JH, Ma LQ, Sun HJ, Cai F, Luo J (2014) Antimony uptake, translocation and speciation in rice plants exposed to antimonite and

- antimonate. *Sci Total Environ* 475:83–89. <https://doi.org/10.1016/j.scitotenv.2013.12.103>
- Sun L, Guo D, Liu K, Meng H, Zheng Y, Yuan F, Zhu G (2019) Levels, sources, and spatial distribution of heavy metals in soils from a typical coal industrial city of Tangshan, China. *Catena* 175:101–109. <https://doi.org/10.1016/j.catena.2018.12.014>
- Tan SY, Praveena SM, Abidin EZ, Cheema MS (2016) A review of heavy metals in indoor dust and its human health-risk implications. *Rev Environ Health* 31(4):447–456. <https://doi.org/10.1515/revveh-2016-0026>
- USEPA., 2006. Integrated Risk Information System (IRIS). Assessments. U.S. Environmental Protection Agency, Washington, DC, USA. Available at: <https://cfpub.epa.gov/ncea/irisdrafts/AtoZ.cfm>
- Vatanpour N, Feizy J, Hedayati Talouki H, Es'haghi Z, Scesi L, Malvandi AM (2020) The high levels of heavy metal accumulation in cultivated rice from the Tajan river basin: health and ecological risk assessment. *Chemosphere* 245:125639. <https://doi.org/10.1016/j.chemosphere.2019.125639>
- Wan M, Hu W, Wang H, Tian K, Huang B (2021) Comprehensive assessment of heavy metal risk in soil-crop systems along the Yangtze River in Nanjing, Southeast China. *Sci Total Environ* 780:146567. <https://doi.org/10.1016/j.scitotenv.2021.146567>
- Wang L, Ma L, Yang Z (2017) Spatial variation and risk assessment of heavy metals in paddy rice from Hunan Province, Southern China. *Int J Environ Sci Technol* 15(7):1561–1572. <https://doi.org/10.1007/s13762-017-1504-y>
- Wang Y, Yang L, Kong L, Liu E, Wang L, Zhu J (2015) Spatial distribution, ecological risk assessment and source identification for heavy metals in surface sediments from Dongping Lake, Shandong, East China. *Catena* 125:200–205. <https://doi.org/10.1016/j.catena.2014.10.023>
- Wang M, Chen W, Peng C (2016) Risk assessment of Cd polluted paddy soils in the industrial and township areas in Hunan, Southern China. *Chemosphere* 144:346–351. <https://doi.org/10.1016/j.chemosphere.2015.09.001>
- Wu W, Wu P, Yang F, Sun DL, Zhang DX, Zhou YK (2018) Assessment of heavy metal pollution and human health risks in urban soils around an electronics manufacturing facility. *Sci Total Environ* 630:53–61. <https://doi.org/10.1016/j.scitotenv.2018.02.183>
- Xiao G, Hu Y, Li N, Yang D (2018) Spatial autocorrelation analysis of monitoring data of heavy metals in rice in China. *Food Control* 89:32–37. <https://doi.org/10.1016/j.foodcont.2018.01.032>
- Yang Z, Jing F, Chen X, Liu W, Guo B, Lin G, Huang R, Liu W (2018) Spatial distribution and sources of seven available heavy metals in the paddy soil of red region in Hunan Province of China. *Environ Monit Assess* 190(10):611. <https://doi.org/10.1007/s10661-018-6995-6>
- Yuanan H, He K, Sun Z, Chen G, Cheng H (2020) Quantitative source apportionment of heavy metal(loid)s in the agricultural soils of an industrializing region and associated model uncertainty. *J Hazard Mater* 391:122244. <https://doi.org/10.1016/j.jhazmat.2020.122244>
- Zeng F, Wei W, Li M, Huang R, Yang F, Duan Y (2015) Heavy metal contamination in rice-producing soils of Hunan province, China and potential health risks. *Int J Environ Res Public Health* 12:15584–15593. <https://doi.org/10.3390/ijerph121215005>
- Zhang Z, Lu Y, Li H, Tu Y, Liu B, Yang Z (2018a) Assessment of heavy metal contamination, distribution and source identification in the sediments from the Zijiang River, China. *Sci Total Environ* 645:235–243. <https://doi.org/10.1016/j.scitotenv.2018.07.026>
- Zhang J, Li H, Zhou Y, Dou L, Cai L, Mo L, You J (2018b) Bioavailability and soil-to-crop transfer of heavy metals in farmland soils: a case study in the Pearl River Delta, South China. *Environ Pollut* 235:710–719. <https://doi.org/10.1016/j.envpol.2017.12.106>
- Zhang Z, Wu X, Wu Q, Huang X, Zhang J, Fang H (2020a) Speciation and accumulation pattern of heavy metals from soil to rice at different growth stages in farmland of southwestern China. *Environ Sci Pollut Res Int* 27:35675–35691. <https://doi.org/10.1007/s11356-020-09711-2>
- Zhang Z, Zhang N, Li H, Lu Y, Yang Z (2020b) Potential health risk assessment for inhabitants posed by heavy metals in rice in Zijiang River basin, Hunan Province, China. *Environ Sci Pollut Res Int* 27(19):24013–24024. <https://doi.org/10.1007/s11356-020-08568-9>
- Zhao K, Liu X, Xu J, Selim HM (2010) Heavy metal contaminations in a soil-rice system: identification of spatial dependence in relation to soil properties of paddy fields. *J Hazard Mater* 181(1-3):778–787. <https://doi.org/10.1016/j.jhazmat.2010.05.081>
- Zhao Q, Wang Y, Cao Y, Chen A, Ren M, Ge Y, Yu Z, Wan S, Hu A, Bo Q, Ruan L, Chen H, Qin S, Chen W, Hu C, Tao F, Xu D, Xu J, Wen L, Li L (2014) Potential health risks of heavy metals in cultivated topsoil and grain, including correlations with human primary liver, lung and gastric cancer, in Anhui province, Eastern China. *Sci Total Environ* 470-471:340–347. <https://doi.org/10.1016/j.scitotenv.2013.09.086>
- Zheng S, Wang Q, Yuan Y, Sun W (2020) Human health risk assessment of heavy metals in soil and food crops in the Pearl River Delta urban agglomeration of China. *Food Chem* 316:126213. <https://doi.org/10.1016/j.foodchem.2020.126213>
- Zhou L, Yang B, Xue N, Li F, Seip HM, Cong X, Yan Y, Liu B, Han B, Li H (2014) Ecological risks and potential sources of heavy metals in agricultural soils from Huanghuai Plain, China. *Environ Sci Pollut Res Int* 21(2):1360–1369. <https://doi.org/10.1007/s11356-013-2023-0>

This manuscript was only submitted on *Environmental Science and Pollution Research*.

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.