Original Research

Characteristics of Cd Contents in Vegetables Around an Abandoned Aluminum Factory and the Potential Health Risks

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Abstract

Wastes and dust discharged from aluminum factory generally result in soil contamination and may cause enrichment of Cadmium (Cd) in local crops. However, the pattern of Cd enrichment in the vegetables by contaminated soil of aluminum factory and their health risk are unclear. In this study, a total of 12 species of vegetables including 163 samples were collected around an abandoned aluminum factory. The results showed that 21% of the leafy vegetables with Cd exceeding Food Safety Standard Limit of China. The Cd in vegetables collected from different distances of four directions from the abandoned aluminum factory had no significant differences (*p*>0.05). Our results indicate that the highest hazard quotient (HQ) and carcinogenic risk of Cd in vegetables are 1 km and 7-10 km away from abandoned aluminum factory respectively. The HQ values of Cd in vegetables for children and adults are within the safe range, however CR values of Cd in vegetables for both children and adults are higher than the acceptable limit of USEPA (10⁻⁶), suggesting potential health risks. The health risks for children aged 2-6 years old are higher than aged 7-17 years old and adults due to the less amount of vegetable intake by children. The intake risks of Cd in different vegetables are spinach>green vegetables>red leafy vegetables>cabbage>radish leafy>stem vegetables>sweet potato leaves>pumpkin leaves>beans.

Keywords: cadmium, vegetable, abandoned aluminum factory, health risk

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Introduction

Cadmium (Cd) is easy to be translocated between environmental media or from environment to organism. It will not only cause environmental pollution, but also cause potential health risks to the population [1]. Most scientists cite the identification of Itai-itai (ouch-ouch) disease in Japan during the 1960s as the first recognition of Cd to health damage [2-4]. Up to now, a large number of epidemiological studies and animal experiments have shown that Cd poisoning could cause a variety of cancers, such as lung cancer, prostate cancer, kidney cancer, liver cancer, etc [5, 6]. The International Agency for Research on Cancer listed Cd as a human carcinogen in 1993 [7]; US National Toxicology Program also pointed out that Cd was a cause of cancer, and classified as Group I in cancerous incidence [8].

Vegetables are one of the indispensable foods in life and the main source of essential trace elements for the human body. Therefore, enrichment of heavy metals in vegetables can cause great damage to human health [9, 10]. Studies show that vegetables contaminated with heavy metals were mainly grown around mines and the use of agricultural fertilizer [11]. Here, affected by the discharge of industrial "three wastes", untreated or unqualified sewage is used for farmland irrigation, and heavy metals in sewage gradually accumulate in the soil, resulting in the accumulation of heavy metals in vegetables [12]. Soil pollutants are mainly heavy metals, and the main causes of pollution derived from the excessive emissions of industrial and mining wastes and smoke [13]. In previous reports, the contaminant Cd is pervasive in China. For example, nearly 100 tons of rice in Hunan province of China was destroyed because of excessive Cd accumulation caused by the longterm environmental pollution of the mining industry. Consequently, health damage caused by Cd has been arousing increasingly concern in China [14, 15].

It is clear that the soil around aluminum factories was generally contaminated by heavy metals, and it is easy to cause enrichment of heavy metals in vegetables [16, 17]. For instance, the maximum value of Cd in the soil around the abandoned aluminum plant in Guizhou of China was 1.4 mg·kg⁻¹, which was 2.2 times higher than the standard limit [18]. Cd pollution was the most serious in the soil around an aluminum factory in Baotou, the average value is 0.60 mg·kg⁻¹, and the overshoot rate was 100% [19], and the spatial distribution of soil heavy metal Cd mainly exists in the surface layer [20]. Studies have shown that the spatial distribution of pollutants in crops around factories is affected by direction, altitude, distance and wind direction [21]. In Shandong Province, the Cd content in vegetables (spinach and rape) around the aluminum factory exceed the National Standard Limit of China [22, 23]. In addition, the abandoned aluminum plant in Guizhou is surrounded by interlaced traffic, dense distribution of population and fields,

and the planting of vegetables is also common. These results indicate that Cd in the soil surrounding the aluminum plant remains a significant ecological risk regardless of whether the aluminum plant is in production or abandoned for many years. If the land adjacent to the abandoned land is converted to agricultural use, even if the aluminum plant is abandoned for many years, there will be a huge potential health risk to the local community. As a result, a rigorous risk assessment is required before an abandoned area can be converted into farmland. Therefore, the main purpose of this study is (1) to explore the content and spatial distribution characteristics of Cd in different vegetables on the basis of previous studies on Cd in soil around aluminum plants; (2) To assess the health risks of the exposed population after ingesting different vegetables.

Materials and Method

Study and Sample Areas

The study area is located around Guizhou Aluminum Factory in Guiyang city, Guizhou Province, China. The aluminum factory is established in 1958 and ceased in August 2014, which is one of the largest electrolytic aluminum plants in China and with an annual output of 1.2 million tons of alumina. It is run for more than 60 years and his numerous industrial wastes were dispersed into surrounding environment. The west of the factory is a residential area, and the east side is dispersedly covered by forest. The annual average temperature is 12.5ºC-14.5ºC and the annual average rainfall is 1147-1191 mm. The outcropped lithology is mainly composed of limestone accompanied by topography of low hills. After self-purified for several years, some local farmers cultivated vegetables in these abandoned lands.

Vegetables Sampling and Analysis

In this study, a total of 12 species of 163 vegetable samples from 5, 8, 9 and 7 sampling areas along the four directions (A: Northeast, B: Northwest, C: Southwest and D: Southeast) surrounding the abandoned aluminum factory were collected (Fig. 1). The studied samples were composed of 141 leafy vegetables (cabbage, sweet potato leaves, spinach, pumpkin leaves, sweet potato leaves and radish vegetables, etc.) and 22 legume vegetables (sword beans and corn).

According to the "China National Food Safety Standard" [24], the samples are processed as follows. Samples were brought back to the laboratory and washed with purified water, then dried in an oven at 60ºC. Dry samples were grinded to 100 mesh and stored in bags. About 0.30 grams of solid was weighed and translocated in the PTFE tube, then 5.0 mL HNO₃ was added and left overnight. The tube was tightly closed with outer tank and placed in an oven at 160ºC

Fig. 1. Study area and sampling distribution in Yunyan district, Guiyang, Guizhou, China.

for 6 hours. The PTFE tube was placed on electric heating plate (100ºC) and heated to nearly dry, then the paste was dissolved with diluted $HNO₃$ and stored in a 50 mL volumetric flask before analyzing.

The Cd content in the digested solution was measured by inductively coupled plasma mass spectrometer (ICP-MS, NexlON2000). In every batch, reagent blanks (4%), reference standards (2%) and duplicate samples (10%) was applied to evaluate the accuracy and precision. The standard reference substance GBW10012 (GSB-3, rice) from China National Standard Material Research Center was used to verify the accuracy of the method, which showed the Cd recovery was 90%-110%.

Health Risk Assessment

In this study, the carcinogenic risk and the hazard quotient (HQ) assessment models were developed based on USEPA, which were used to evaluate the health risks of Cd in vegetables [25]. The calculation formulas are shown in Table 1, and key parameters are displayed in Table 2. Where CDI $(mg (kg \cdot d)^{-1})$ represents the average daily intake of Cd into the human body through vegetables; RfD is the dietary reference dose of heavy metals recommended by USEPA $(1\times10^{-3} \text{ mg} \cdot (\text{kg} \cdot \text{d})^{-1})$ [25]: HO is the single health risk of non-carcinogenic index of Cd. When HQ value is lower than 1, it is considered as the acceptable level for human health risk; CR is the single health index of carcinogenic risk of Cd; SF is the slope coefficient of carcinogenic of Cd through food intake $(6.1 \text{ (kg d) mg}^1)$ [25]; CR values less than 10-6 could be considered as safe level, and greater than 10-4 means that attention should be paid.

In this study, health risk assessment was conducted on the median P_{50} (50th percentile) of heavy metal contents in vegetables. The assessment results are limited to point estimates, and can only represent the health risk status of the middle-level population. Therefore, the uncertainty is introduced that the 10th percentile (P_{10}) of the sample is determined as the lower limit of uncertainty, and the 90th percentile (P_{00}) of the sample is the upper limit of uncertainty. The safety limit of HQ and ILCR should below 1×10-4 respectively [28].

Statistical Analysis

SPSS was used to complete descriptive statistics, variation coefficient and Spearman rank correlation. Because all of the data were non-normally distributed, the significant differences in the Cd concentrations of vegetables were analyzed by the Mann-Whitney U test. The Spearman rank correlations were performed to determine the relationships between the Cd exposure of direction and distance.

Table 1. Calculation formula of health risk assessment model.

Type of risk	Calculation formula			
Chronic Daily Intake	$MC \times IR \times EF \times ED \times Fd$ $CDI =$ $BW \times AT$			
Non-carcinogenic risk	CDI $\overline{\text{RfD}}$ (2)			
Carcinogenic risk	$CR = CDI \times SF(3)$			

Parameter	Mean	Adult	Child (6-17 years)	Child (2-6 years)	References
BW	Weight/kg	60	39.63	16.61	$[26]$
IR.	Intake rate $/(kg \cdot d^{-1})$	0.402	0.183	0.132	
ED	Exposure Day /a	54	62	70	$[25]$
EF	Exposure Frequency $/d \cdot a^{-1}$	365	365	365	$[25]$
Fd	The proportion of fresh weight converted to dry weight	0.1	0.1	0.1	$[27]$
MC.	Median Cd content /(mg·kg-1)				
AT	Average contact time/d	$ED\times365$	$ED\times365$	$ED\times365$	$[25]$

Table 2. Exposure parameters of health risk assessment.

Results and Discussion

Pollution Concentration of Cd in Vegetables

The average Cd concentrations of leafy vegetables and legumes are 0.035 mg·kg⁻¹ and 0.014 mg·kg⁻¹, respectively. The average Cd concentrations are significantly different between two groups (*p*<0.01), which suggested that leafy vegetables have higher accumulate capacity of Cd than beans. Our result indicates that Cd in about 21% of leafy vegetables and none beans exceeded the China National Food Safety Standard (CNFSS) (Table 3). The 90th quantile is 0.08 mg kg¹, which indicates potential risks for local residents. Research results showed that the

highest cadmium content in spinach around Donga Aluminum Plant was 0.049 mg·kg-1, with an average of 0.031 mg·kg⁻¹ (measured by fresh weight) [29]. This is similar to the results of this study. The cadmium uptake and accumulation of crops from high to low were roots, stems, leaves, ears and seeds, among which, the accumulation of cadmium in roots accounted for 50%~80%, while the cadmium in seeds only accounted for a small part of the total amount of cadmium in plants, and distributed evenly in seed coat and endosperm [30, 31]. Therefore, the cadmium content in stem and leaf vegetables is higher than that in fruit vegetables.

Taking the aluminum plant as the center, there was no statistically difference in the cadmium content

Fig. 2. Box diagram analysis of cadmium concentration in vegetables around the aluminum plant in different directions and distances.

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 \top T of vegetables in the four directions (*p*>0.05). Among them, the Northeast (NE) direction had the highest concentration of cadmium in vegetables, with an overstandard rate of 27%, and the lowest Cd excess rate in vegetables in the northwest was 13%. This may suggest that wind direction was not the main factor affecting cadmium pollution around aluminum plants (Fig. 2a).

The analysis of the distance from the aluminum factory shows that the vegetables have the highest content of cadmium at 1 km and 7-10 km from the aluminum factory. It can be found from Fig. 2b) that the cadmium content in vegetables had a downward trend with the increase of distance in 6 km. When the distance exceeded 6 km, the Cd content in vegetables began to increase again, and the difference in the degree of cadmium enrichment between distances was statistically significant (*p*<0.05). The distance to the mining area is not the main factor affecting soil heavy metal pollution, but may also be related to factors such as topography, slope direction and wind direction [10]. For large-scale smelting enterprises, the distance of artificial diffusion halo can reach 25-30 km, and the content of metal pollutants in plants is powerdependently reduced from the pollution source to the background area [32, 33].

Source Analysis

Correlation Analysis

The correlation between the Cd content of different varieties of vegetables and the planting altitude is shown in (Fig. 3). After fitting the scatter plot, it is found that the correlation between the total vegetable samples and planting altitude is not significant $(r = -0.07, p = 0.35)$. Among them, the Cd content in cabbage, beans, pumpkin leaves and green vegetables (Fig. 3a, Fig. 3b, Fig. 3d, and Fig. 3e) showed a decreasing trend to varying degrees with the increase in altitude. As the slope increased, heavy metal contents increased then decreased [34, 35]. Cd content in sweet potato leaves, stem vegetables and radish leaves (Fig. 3c, Fig. 3f and Fig. 3g) showed a different degree of upward trend with the elevation. The correlation analysis between the Cd content in vegetables and the distance from the aluminum plant of the four directions is shown in (Fig. 4). After fitting the scatter plot, it is found that the cadmium content in vegetables increases with distance in the NE, SW and SE directions. There is an upward trend in varying degrees, but the opposite is true in the NW direction. Wind directions have a great influence on the distribution of metals in coal, gold, and fluorite mine area. The concentration of heavy metals in mine area increased with the distance from the center of the mine, then reached a peak, then decreased gradually [36]. In this study, the results obtained are somewhat different from these studies. In theory, manmade environmental geochemical anomalies should be concentric circles, and the concentration of diffused

elements gradually decreases from the pollution source to the surrounding area [37, 38]. But in fact, concentric circles are often destroyed under the influence of the prevailing wind direction and atmospheric precipitation [39, 40]. The width of the pollution zone changes drastically. The air masses carrying aerosols are also affected by the terrain when they move. Therefore, the anthropogenic geochemical anomalies of heavy metals may be extended or shortened and have irregular contours. This also explains the irregular distribution characteristics of cadmium content in soil around the abandoned aluminum plant in Guizhou, resulting in no significant correlation between cadmium content in vegetables and altitude.

Influence Factor

The Cd enrichment of vegetable samples in different directions didn't reach significant level, but the Cd content of P_{50} in the NE direction was the largest, with a value of 0.031 mg·kg⁻¹. The content of Cd in vegetables is relatively higher in the range of 1 km and 7-10 km from the abandoned aluminum factory. The lowest area in the research region locate at 7-10 km northeast of the abandoned aluminum factory, with an altitude of 1172 m (Fig. 5). Besides, the perennial wind direction is southeast and northeast wind (Fig. 6), which may be the main reasons for the highest content of Cd in vegetables in this direction. The possible reasons for this result are that the waste or smoke generally deposit on the low-lying areas. Some reports found the source and the distance to the mining area were not the main factor affecting soil heavy metal pollution, but may also be related to factors such as topography, slope direction and wind direction [10, 34, 41]. Cadmium pollution in aluminum plant production can be divided into three parts (waste gas, waste residue and waste water), and the pollution range of waste residue and waste water is greatly affected by human factors. However, meteorological factors may directly affect the precipitation of cadmium-containing soot and aerosols in the exhaust gas, leading to more serious cadmium pollution in perennial wind direction. In this wind direction, the natural factors of wind may carry cadmium-containing soot to distant soil, resulting in soil cadmium pollution, and cadmium is mainly absorbed by plants through soil.

Potential Health Assessment

From the perspective of different directions, distance range and total samples, non-carcinogenic and carcinogenic health risks of cadmium content in cultivated vegetables were assessed (box image plot and normal curve), as shown in (Fig. 7). As can be seen from the normal curve, the distribution of HQ and CR of cadmium in vegetables is skewed, so the median (the median line of the box plot) is used to describe the results. Fig. A, B and C are the non-carcinogenic

Fig. 3. Correlation between Cd content and altitude in different vegetables around the abandoned aluminum factory.

health risk (HQ) evaluation. As shown in the figure, the non-carcinogenic risk (HQ adults = 0.01, HQ children $(7-17 \text{ years}) = 0.01$, HQ children $(2-6 \text{ years}) = 0.02$ caused by cadmium in total vegetable samples to local residents are all less than 1. But the non-carcinogenic risk is highest in children aged 2-6. From the direction of aluminum plant, NE direction has the highest risk (HQ children (2-6 years) = 0.025, HQ children (7-17 years) = 0.01 , HQ adult = 0.02), NW direction has the lowest risk; from the distance to the aluminum plant, the non-carcinogenic risk of cadmium in vegetables from 2-3km away from the aluminum plant (HQ children (2-6 years) = 0.03 , HQ children (7-17 years) = 0.02 , HO adults = 0.02) is the largest, and the minimum from 4-6km away. (Fig. 7d, e and f) are the evaluation of carcinogenic risk [42]. The results showed that in total vegetable samples, CR children aged (2-6 years) $= 1.0 \times 10^{-4}$, CR children aged (7-17 years) $= 0.5 \times 10^{-4}$, and CR adults = 0.8×10^{-4} . NE direction has the highest risk of cancer (CR children (2-6 years) = 1.5×10^{-4} , CR children (7-17 years) = 0.8×10^{-4} , CR adults = 1.3×10^{-4}). The range of 2-3km has the highest risk of cancer (CR children (2-6years) = 1.7×10^{-4} , CR children (7-17years) $= 0.9 \times 10^{-4}$, CR adults $= 1.5 \times 10^{-4}$). Cancer risk is greater than 10^{-4} in children (2-6 years) and 10^{-6} in children (7-17 years) and adults. These associated risks deserve attention to ensure the safety and health of children.

The Cd enrichment degree in different vegetables (cabbage, beans, sweet potato leaves, etc.) around the abandoned aluminum factory was statistically

Fig. 4. Correlation between Cd content and radius of vegetables around the abandoned aluminum factor.

Fig. 5. Altitude cross-sections of abandoned aluminum plants in different directions [21].

Fig. 6. Perennial wind direction in Guizhou Province.

Fig. 7. Non-carcinogenic and carcinogenic health risk assessment of Cd in vegetables around the abandoned aluminum plant to people with different characteristics.

described in Table 4. From the median of Cd in all samples, spinach>green vegetables>red leafy vegetables >cabbage >radish leafy>stem vegetables>sweet potato leaves>pumpkin leaves>beans, among which spinach has the highest value of 0.06 mg·kg⁻¹. From the $90th$ quantile point of view, cabbage $(0.09 \text{ mg} \cdot \text{kg}^{-1})$, stem vegetables (0.07 mg·kg-1), radish leaves (0.10mg·kg-1) and green cabbage $(0.16 \text{ mg} \cdot \text{kg}^{-1})$, which exceed the CNFSS Limit standard (0.05 mg·kg-1). From the health risks of children and adults, HQ (Children 2-6 years old)>HQ (Adults)>HQ (Children 7-17 years old), CR (Children 2-6 years old)>CR (Adults)>CR (Children 7-17 Years old); Cd has the greatest health risk in spinach, HQ (Adults) = 0.04, HQ (Children 7-17 years old) = 0.03 , HQ (Children 2-6 years old) $= 0.05$; CR (Adults) $= 2.4 \times 10^{-4}$, CR (Children 7-17 years old) = 1.6×10^{-4} , CR(Children 2-6 years old) = 2.9×10^{-4} . The carcinogenic risk exceeds the safety limit (10⁻⁴) 2.4, 1.6 and 2.9 times, respectively. The carcinogenic risk of sweet potato leaves was the lowest, which was within the safety limit set by the USEPA. For children (2-6 years old), only beans vegetables (CR = 0.34×10^{-4}), sweet potato leaves ($CR = 0.34 \times 10^{-4}$) and pumpkin leaves (0.39×10^{-4}) < 1×10⁻⁴. However, other types of vegetables have cancer risk. For children (7-17 years old), CR (red leafy vegetables) and CR (spinach) $>1\times10^{-4}$. For adults, CR (cabbage), CR (red leafy vegetables), CR (radish leaves), CR (green vegetables) and CR (spinach) $>1\times10^{-4}$. Previous studies found that vegetables enriched of Cd in the order of spinach>Chinese cabbage>radish [43, 44], which is consistent with our results. Because of the species difference among different plants, the ability of cadmium enrichment from soil is also different, that is, the bioconcentration factor (BFC) of cadmium is different among different vegetables, leading to the different content of cadmium in different vegetables [45]. The Cd in the total vegetable samples has no non-carcinogenic risk, but the carcinogenic risk factor exceeds the acceptable safety limit of 10^{-6} , suggested potential carcinogenic risk.

There were researches discussed the health risks of heavy metals in soil and vegetables in mining areas to the human body, they found that the health risks caused by children are higher than those of adults, which are different from the results of this study [20]. The reason is that health risk assessment is not only relate to the degree of enrichment and potential harm of heavy metals in vegetables, as well as weight coefficient and food intake amount. This study focuses on comparing the health risks of children 2-6 years old, children 7-17 years old and adults.

This study aims to understand the degree of cadmium accumulation in vegetables around abandoned aluminum plants in Guizhou, to explore the factors affecting cadmium accumulation in vegetables, and to assess the health risks of oral intake by children and adults. However, the evaluation of the health risks of single-factor Cd is more limited. In order to more accurately understand the health risks caused by

heavy metals, the combined effect of multiple factors and multiple exposure factors should be further considered [46, 47]. In areas that are heavily polluted by Cd, local residents should be encouraged to grow beans and vegetables, or in heavily polluted areas can be forested through land, through a long-term natural purification process to eliminate high levels of heavy metals in the soil.

Conclusion

The vegetables around the abandoned aluminum factory showed significant enrichment of Cd. Affected by the geomorphic and meteorological conditions, the Cd contents in vegetables were not significantly related to the direction and the distance from the abandoned aluminum factory. This may be mainly influenced by the soil environment

The CR values for non-carcinogenic risks of vegetables in the studied area were all in the scope of the safety range, but the carcinogenic risk exceeded the safety limit, which indicated that there was a potential carcinogenic risk. Among different vegetable species, spinach has the highest enrichment of Cd, and legumes have the lowest enrichment. Due to effect of vegetable intake amount, the health risks of Cd in vegetables for children 2-6 years old were higher than children 6-17 years old and adults.

The observation on the enrichment of Cd in collected vegetables indicated that the impact of abandoned aluminum factory on the surrounding environment exist persistently for many years, stringent risk evaluation is necessary before these abandoned area converts to agricultural field.

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Conflict of Interest

The authors declare no conflict of interest.

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