(cc) BY

Distribution and health risk evaluation of heavy metal lead in the main production area of rice in Heilongjiang Province

Xinhui WANG^{1#} (D, Xuejian SONG^{1,2,3#}, Dongmei CAO^{1,2,3*} (D, Dongjie ZHANG^{1,2,3*} (D, Zhijiang LI^{1,2,3}, Chang ZHANG¹

Abstract

In order to explore the distribution and transfer of lead elements in soil and rice in the five regions of Chahayang, Wuchang, Fangzheng, Xiangshui and Jiansanjiang in Heilongjiang Province, and analyze the impact of rice intake on human health, the samples were tested by ICP-MS. Modeling the lead-element transfer in the soil-rice system, using the Nemero comprehensive pollution index method and the health risk assessment model to evaluate the lead pollution status of rice in the study area and its health risks to adults and children. The results showed that the average content of lead in rice in the study area was Chahayang 0.02 mg/kg, Wuchang 0.03 mg/kg, Fangzheng 0.017 mg/kg, Xiangshui 0.023 mg/kg and Jiansanjiang. 0.024 mg/kg did not exceed the lead content limit specified by China's National Food Hygiene Standard (0.2 mg/kg); Based on the prediction model of heavy metal lead content, pH value, and organic matter in the soil, the transfer of lead elements in the soil-rice system of Chahayang, Fangzheng, Xiangshui, and Jiansanjiang rice fields can be significantly described, with R² values ranging from 0.224 to 0.419; Both the pollution index and the comprehensive pollution index in the study area were less than 1, which belong to the non-pollution category. The health risk index of heavy metal lead for adults and children in all five regions is lower than the maximum acceptable risk level recommended by USEPA, and there is no risk of causing cancer.

Keywords: soil; rice; lead; pollution characteristics; health risk.

Practical Application: The predictive model developed in this study can significantly describe the transfer of lead in the "soil-rice" system in rice fields.

1 Introduction

Lead is a heavy metal element that is very toxic to pollutants. It has a large toxic effect on plants and animals, and has adverse effects on plant morphology, growth, and photosynthesis (Cao et al., 2015). The interaction of lead with biomolecules damages the animal's reproductive, nervous, immune, cardiovascular and other systems, affecting growth and development. Similarly, lead also causes great harm to human health. After entering the body, lead is mainly accumulated in bones, arteries, liver, kidneys, pancreas and lungs, and can also enter the brain (Singh et al., 2003; Mushak, 2003). Lead can replace calcium in bone and store in bone. It can cause damage to many organs and systems such as the central and peripheral nervous system, blood system, kidney, cardiovascular system and reproductive system, and can cause cognitive abilities and behaviors. Functional changes, genetic material damage, apoptosis induction, etc., and have certain mutagenicity and carcinogenicity (Silbergeld et al., 2000; Fang et al., 2014). The normal upper limit of urine lead in China is 0.08 mg/L. Even if a very low amount of lead is taken daily, it will accumulate in the body and cause chronic poisoning and even carcinogenesis. Lead is more harmful to children and is a strong neurophilic poison that affects children's intelligence, behavior, ability, and normal growth and development. Lead can also increase the incidence of infants with congenital defects (Simon et al., 2007).

As a country with a large population, agriculture is the primary industry. Most people in the country rely on rice as the main food (Wei & Yang, 2010). With the development of social industrialized economy, the problem of heavy metal pollution of crops has become a region of great concern. Scholars have done a lot of research on the potential risks of heavy metals to human health. However, their research fields are mostly concentrated in mining areas (Yan et al., 2022; Cai et al., 2015), sewage irrigation areas (Wang et al., 2017; Zeng et al., 2015), and waste treatment plants (Liu et al., 2021; Li et al., 2015) and other places. The research objects focus on dust reduction (Luo et al., 2022; Mehr et al., 2016), corn (Salam et al., 2022; Yu et al., 2017), Vegetables (Jalali & Meyari, 2022; Chang et al., 2014), drinking water (Giri & Singh, 2015; Pan et al., 2022), rice (Zhang et al., 2022; Lu et al., 2022), and so on. These studies have conducted in-depth research on the accumulation characteristics, physical and chemical properties, occurrence forms of heavy metals, and the physiological and

Received 03 Jan., 2023

Accepted 30 Jan., 2023

¹College of Food Science, Heilongjiang Bayi Agricultural University, Daqing, Heilongjiang Province, China

²Key Laboratory of Agro-Products Processing and Quality Safety of Heilongjiang Province, Daqing, Heilongjiang Province, China

³National Coarse Cereals Engineering Research Center, Daqing, Heilongjiang Province, China

^{*}Corresponding author: caodong3018@sina.com; byndzdj@126.com

^{*}Xinhui WANG and Xuejian SONG contributed equally to this work and should be regarded as co-first author.

biochemical effects of polluted bodies after pollution, laying a foundation for further research on the accumulation of heavy metals in crops and their effects on human health. However, most of these studies are pot experiments under artificially controlled conditions, but not many studies have been conducted on the accumulation characteristics of heavy metals in rice grown in the natural environment. The concept of risk analysis and risk assessment first came to the attention of the people of China. The 2015 revision of the Food Safety Law of the People's Republic of China clearly defines the responsibilities and personnel related to risk assessment in each department. So far, China has collected nearly 300 monitoring indicators of food biological, physical and chemical viral hazards in 30 major categories of food every year, preliminarily designed and established a national foodborne viral disease monitoring database, and collected relevant data of various food production and processing, persistent organic chemical pollutants, fungi and viruses, heavy metal pollutants, etc.

In recent years, with the rapid development of economy and the acceleration of industrialization and urbanization, the growth environment of agricultural products such as agricultural land and irrigation water has been seriously polluted. Heavy metal pollution incidents occur frequently, affecting environmental resources, agricultural development and people's health. This article studies the content of heavy metal lead in soil and rice and its harm to human health. The soil and rice in some regions of Heilongjiang, the largest grain silo in China, were selected for testing to understand the pollution status and content of heavy metal lead. People in the five regions of Chahayang, Jiansanjiang, Fangzheng and Xiangshui passed the risk assessment of lead ingestion of rice in the hope of understanding the specific situation of lead pollution and the potential health caused by rice consumed by residents influences. Analysis of the pollution situation can alert us to sustainable development, not just focusing on gross national product, but also focusing on environmental protection and human health.

2 Research method

2.1 Overview of the study area

Heilongjiang Province is located in the rice-growing area with the northernmost latitude. It has large temperature difference between day and night, fertile soil, excellent water quality and low pollution. It is conducive to the development of rice production. Heilongjiang rice production is now entering a new era of high quality, high efficiency and professionalism. Due to stable production, excellent rice quality and high commodity rate, Heilongjiang rice has become an important high-quality glutinous rice production base in China, and its products are shipped to all parts of the country. In 2015, rice planting area in Heilongjiang Province accounted for 17.0% of the country's rice planting area, and represented 69.25% of the rice planting area in the three northeastern provinces. Rice production in Heilongjiang accounts for 16.3% of the national rice production, and 67.6% of the rice production in the three northeastern provinces, playing an important role in the rice market.

2.2 Sample source

The rice and corresponding soil samples used in this study were from the rice producing areas in Heilongjiang province, including 22 from Chahayang region, 22 from Wuchang region, 22 from Fangzheng region, 22 from Xiangshui region, and 22 from Jiansanjiang region.

2.3 Sample testing

The determination of the content of Pb in rice samples was carried out according to the method specified in Chinese National Standard GB 5009.286-2016 "Determination of Multi-Elements in Foods".

2.4 Risk assessment of heavy metal pollution in rice grains

The limit of Pb in rice grains was based on the limits of 8 elements including lead, chromium, cadmium, mercury, selenium, arsenic, copper and zinc in grain (including cereals, beans and potatoes) and products (NY861-2004). The single factor pollution index method and the Nemero comprehensive pollution index method were used to evaluate the heavy metal content in crops (Baker et al., 1994).

Single factor pollution index:

$${}^{P}_{Pb} = \frac{C_{Pb}}{S_{Pb}}$$
(1)

In the formula, $P_{\rm Pb}$ is the comprehensive pollution index of heavy metals in crop grains; $C_{\rm Pb}$ is the average value of single metal pollution index of heavy metals, and $S_{\rm Pb}$ is the maximum value of one-way pollution index of heavy metal Pb.

Nemero Integrated Pollution Index:

$$P = \sqrt{\frac{\overline{P}_{Pb}^{2} + P_{Pb} \max^{2}}{2}}$$
(2)

In the formula, P is the comprehensive pollution index of heavy metals in crop grains; \overline{P} is the average value of single metal pollution index of heavy metals, and P_{Pbmax} is the maximum value of one-way pollution index of heavy metal Pb.

2.5 Rice health risk assessment

In order to evaluate the health risks of rice in the diet of adults and children in the study area, the US Environmental Protection Agency (USEPA) recommended health risk assessment model was used (Means, 1989). The model used in this study was the carcinogenic risk model.

Average daily intake of pollutants through crops (ADD) (Formula 3):

$$ADD = \frac{C_{pb} * I * EF * ED}{BW * AT}$$
(3)

where *ADD* is the average daily intake of pollutants in crops $[mg\cdot(kg\cdot d)^{-1}]$; C_{pb} is the content of heavy metal Pb in crops

(mg·kg⁻¹); *I* is the daily human body intake of crops (kg·d⁻¹); *EF* is the frequency of exposure (d·a⁻¹); *ED* is the exposure time (a); *BW* is the recipient body weight (kg); *AT* is the life expectancy (d). The names and values of various parameters are shown in Table 1.

Non-carcinogenic risk assessment.

$$HQ = \frac{ADD}{RfD}$$
(4)

In the Formula 4, *HQ* is the health risk index of heavy metal Pb; *RfD* is the reference measurement of heavy metal exposure [mg·(kg·d)⁻¹]. When $HQ \le 1$, heavy metal pollutants will not cause health risks to the human body; when HQ > 1, it means that heavy metal pollutants will cause health risks to the human body; the greater the HQ, the higher the health risks.

3 Results and analysis

3.1 Lead in soil and rice in Heilongjiang region

The lead content in the 110 soil samples collected was between 19.409 and 33.292 mg/kg, with an average value of 24.287 mg/kg and a coefficient of variation of 0.085% <11%. Within the range of coefficient of variation allowed by GB/T 27404-2008 "Specification for laboratory quality control physical and chemical testing of food", according to GB 15618-2018 "Soil Environmental Quality - Standard for Soil Pollution Risk Control of Agricultural Land (Trial)". Comparison of the content of lead in the soil in the five regions did not exceed the prescribed limit, and the risk of lead pollution from agricultural land in the study area was low. The content of lead in the soil is arranged in descending order: Xiangshui> Chahayang> Jiansanjiang> Wuchang> Fangzheng. The content of lead in brown rice and polished rice in 110 rice samples collected was between 0.002~0.107 mg/kg and 1.8716×10⁻⁵~0.0721 mg/kg, with average values of 0.023 mg/kg and 0.0085 mg/kg, respectively. The coefficients of variation are 0.690% and 1.260%, which are within the range of coefficients of variation allowed by GB/T 27404-2008, and are determined based on the limit standard for lead in rice in GB 2762-2017 "National standard for food safety - limit of pollutants in food" not more than 0.2 mg/kg. The 110 rice samples tested did not exceed this limit, that is, the pass rate was 100%. The content of lead in brown rice is arranged in descending order: Wuchang> Jiansanjiang> Xiangshui> Chahayang> Fangzheng. The content of lead in polished rice is the same as that in brown rice except Chahayang and Jiansanjiang Consistent, far lower than the lead content in rice in Zhejiang

(Zhao et al., 2009). Because the rice produced in the study area is China's national geographical protection mark rice, the production area is located in the Songnen Plain and Sanjiang Plain of Northeast China, and is less affected by urban human activities and mining metallurgy activities. The content does not exceed the standard compliance rate of 100%.

The average content of lead in the soil in this experiment was 24.287 mg/kg, which did not exceed the limit set in the soil environmental quality in China, which was higher than the lead content of 22.00 mg/kg in Heilongjiang soil reported in 2012 (Xia et al., 2014), the content of lead in the soil of Wuchang area was 23.603, which was lower than (Wang et al., 2011) the content of lead in the soil of Harbin area reported in 2011 was 24.6 mg/kg, and The lead content is higher than the other four regions, but the lead content in the soil is not much different. Some studies have shown that the absorption and accumulation of heavy metals by rice are greatly affected by genetic background, variety types, and heavy metal interactions (Srivastava et al., 2014); Some scholars have found that cadmium in the soil will affect rice plants and limit the absorption of lead (Zeng et al., 2008); the single rice variety (rice flower fragrance) and soil background in Wuchang area are the main factors that cause Wuchang and other areas to differ greatly. In the process of absorption and accumulation of heavy metals, rice is not only affected by the heavy metal content in the soil, but also by other factors, such as rice varieties, soil microbial content, precipitation, and air quality (Tables 2, 3 and 4).

Significant analysis of differences between different regions. The SPSS stastistics 12.0 was used to analyze the variance of polished rice, brown rice and soil in five regions. The results are shown in the figure below.

From Figure 1, it can be seen that the contents of lead elements in the soil of Chahayang, Wuchang, and Fangzheng are significantly different; the contents of lead elements in the soil of Wuchang and Xiangshui are significantly different. There was no significant difference in the content of lead elements in the soils of Chahayang, Xiangshui, and Jiansanjiang; there was no significant difference between Wuchang, Fangzheng and Jiansanjiang.

It can be seen from Figure 2 that there is a significant difference in lead content in brown rice between Wuchang and Fangzheng. There was no significant difference in lead content in brown rice between Chahayang, Wuchang, Xiangshui, and Jiansanjiang; there was no significant difference in lead content

Table 1. Parameters of cereal crop health risk assessment model.

M. 11.	Parameter name	Reference		T 't autom	
Model parameter		Adult	Child	Literature	
I/kg·d⁻¹	Intake	0.5	0.177	Duan et al. (2015)	
EF/d·a ⁻¹	Exposure frequency	365	365	U.S. Environmental Protection Agency (1986)	
ED/a	Exposure time	30	10	Means (1989)	
BW/kg	Receptor weight	70	16	U.S. Environmental Protection Agency (1986)	
AT/d	Life expectation	10950	3650	U.S. Environmental Protection Agency (1986)	
RfD/mg·(kg·d)-1	Reference dose		0.0035	Lead and compounds (inorganic); CASRN 7439-92-1	

Study area	Pb				
Study area	Mean ± SD	Range	C·V/%		
Chahayang	24.960 ± 1.547	22.250-26.964	0.162		
Wuchang	23.603 ± 1.663	20.728-27.241	0.151		
Fangzheng	23.096 ± 1.782	19.409-27.531	0.111		
Xiangshui	25.102 ± 2.811	21.335-33.292	0.143		
Jiansanjiang	24.675 ± 1.677	20.909-28.545	0.080		
Total	24.287 ± 2.066	19.409-33.292	0.085		

Table 2. Soil Pb content in different areas (mg kg⁻¹).

Note: C·V is the coefficient of variation.

Table 3. Pb content in milled rice in different areas (mg kg⁻¹).

Study area	Pb			
Study area	Mean ± SD	Range	C·V/%	
Chahayang	0.020 ± 0.009	0.009-0.049	0.434	
Wuchang	0.030 ± 0.018	0.013-0.094	0.599	
Fangzheng	0.017 ± 0.024	0.002-0.107	1.420	
Xiangshui	0.023 ± 0.010	0.011-0.047	0.432	
Jiansanjiang	0.024 ± 0.011	0.011-0.055	0.464	
Total	0.023 ± 0.016	0.002-0.107	0.690	

Table 4. Pb content of refined rice in different regions (mg kg⁻¹).

Study ana	Pb			
Study area	Mean±SD	Range	C·V/%	
Chahayang	0.0080 ± 0.0087	0.0001-0.0334	1.0957	
Wuchang	0.0139 ± 0.1037	0.0001-0.0413	0.7456	
Fangzheng	0.0061 ± 0.0155	1.87×10 ⁻⁵ -0.0721	2.5331	
Xiangshui	0.0079 ± 0.0103	0.0001-0.0355	1.3112	
Jiansanjiang	0.0067 ± 0.0059	0.0001-0.0227	0.8905	
Total	0.0085 ± 0.0107	1.8716×10 ⁻⁵ -0.0721	1.2596	

in brown rice between Fangzheng and Chahayang, Xiangshui and Jiansanjiang.

It can be seen from Figure 3 that there is a significant difference in lead content in polished rice between Wuchang, Fangzheng and Jiansanjiang. There was no significant difference in lead content in polished rice between Chahayang, Wuchang and Xiangshui, there was no significant difference in lead content in polished rice between Fangzheng and Jiansanjiang, Chahayang and Xiangshui.

In summary, the differences in the content of lead in the soil, brown rice, and polished rice in the study area are not consistent. The variety of rice has an effect on the effect of rice on the absorption and accumulation of lead from the soil (Lee, et al., 2016), In addition, some artificial factors such as pesticides, fertilization, and irrigation will greatly affect the absorption of lead by rice. Natural conditions such as precipitation and CO_2 concentration will also affect the absorption and accumulation of lead in rice.

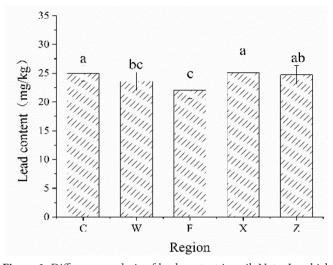


Figure 1. Difference analysis of lead content in soil. Note: In which C is Chahayang, W is Wuchang, F is Fangzheng, X is Xiangshui, J is Jiansanjiang. The same below. Lowercase letters a~c indicate significant differences, the same letters indicate insignificant differences, and different letters indicate significant differences, the same below.

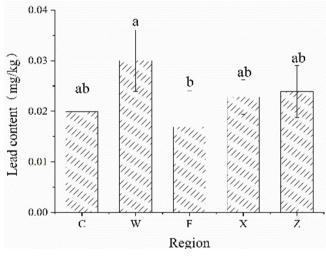


Figure 2. Analysis of Differences in lead Content in Brown Rice.

3.2 Pb element migration model in soil-rice system

The absorption and accumulation of heavy metals in rice is not only affected by the total metal content in the soil, but also by the physical, chemical, and biological characteristics of the soil. Many researchers have studied the factors that affect the absorption of metal elements by rice, including soil pH (Reddy & Patrick, 1977; Dutta et al., 1989), organic matter (Haldar & Mandal, 1979; Kashem & Singh, 2001), redox potential (Sajwan & Lindsay, 1986), salinity (McLaughlin et al., 1996), phosphorus content (Haldar & Mandal, 1981). Studies have found that soil pH is an important factor controlling the absorption of heavy metals (Jung & Thornton, 1997; Basta et al., 2005). Table 5 shows the soil physical and chemical properties in the study area.

Based on previous studies, the factors affecting lead absorption in the "soil-rice system" were studied, and a best-fit model for predicting lead content in rice was proposed. Multivariate regression analysis of lead content in rice was performed using soil pH and organic matter, and a multiple regression model for lead content in rice was established. The multiple regression equation is shown in Table 6. The content of lead in Chahayang rice was significantly negatively correlated with the concentration of lead in soil (P < 0.05); The content of lead in Fangzheng rice showed a significant positive correlation with soil pH (P<0.05), and a significant positive correlation with the concentration of lead in soil (P<0.05); The content of lead in Xiangshui rice was significantly positively correlated with soil pH (P<0.05); The content of lead in Jiansanjiang rice had a significant positive correlation with soil pH (P<0.05). Comparing the content of lead in rice, the concentration of lead in soil, and pH by R² can well predict the content of lead in rice in Chahayang, Fangzheng, Xiangshui and Jiansanjiang areas. However, in the established regression model, lead content in rice and lead content in soil and pH and organic matter values in Wuchang area are not significantly correlated, so the prediction model in this area remains to be studied. Dudka et al. (1996) reported that the relationship between rice and heavy metals in soil can be described by three models: linear model (constant distribution model), plateau model (saturation model), and Langmuir model. The adsorption model may also appear in the range of lower metal concentration in the soil. In this study, through comparing and comparing the three models, it is found that the linear model is the best fit model, so the linear model is used for fitting modeling in this paper. The R² value of the fitted model is between 0.256 and 0.468 (Table 5), and the D-W indices are close to 2. The autocorrelation of the independent variables is not obvious. However, in previous studies, higher fitting coefficients have been

Table 5. Physical and chemical	properties of soil (mean \pm SD).
--------------------------------	-------------------------------------

Study area	pH(range)	Organic matter content(%)	
Chahayang	5.06-8.33	4.02 ± 0.60	
Wuchang	5.37-7.90	3.07 ± 1.22	
Fangzheng	5.33-6.99	3.01 ± 0.93	
Xiangshui	5.57-7.42	3.09 ± 0.93	
Jiansanjiang	5.12-6.68	3.75 ± 0.83	

reported Dudka et al., (1996) reported that the R² values of the correlation between Cd and Zn content in barley grains and Cd and Zn content in soil were 0.94 and 0.92, respectively. McBride (2002) found similar correlation coefficients. The correlation coefficient of this study is lower than the correlation coefficient of previous studies (<0.9). The above-mentioned studies were carried out under the conditions of pot experiments or small test fields, so the soil properties changed little during the modeling process. Within the controllable range, the model established under this condition is less affected by factors and has a higher degree of fit. This experiment was conducted under natural conditions. There is no artificial controllable factor. Paddy soil is a complex system. In addition to the total soil metal content and pH value, other soil properties may play a role in affecting the availability of heavy metals effect. This weakens the ability of the soil metal and pH models to fit.

3.3 Evaluation of lead pollution characteristics in rice grains and health risk assessment of intake

According to the Formulas 1 and 2, the evaluation results of heavy metal pollution in the study area are obtained (Tables 7 and 8). From the single-factor pollution index evaluation results (Table 7), the individual health risks of Pb in the five areas studied The indexes are all less than 1, indicating that these five regions are

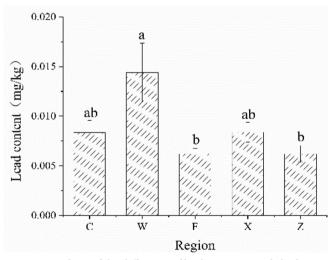


Figure 3. Analysis of the difference of lead content in polished rice.

Table 6. Correlation models for heavy metals in paddy soil rice system in Heilongjiang province.

Study area	N. 11	\mathbb{R}^2	Partial correlation coefficient	
	Model	K-	Metal in soil	Soil pH
Chahayang	$Pb_{rice} = 0.003Pb_{pH} - 0.002Pb_{soil} - 0.002Pb_{OM} + 0.029$	0.239	-0.269*	0.301
Wuchang	$Pb_{rice} = 0.010Pb_{pH} + 0.002Pb_{soil} + 0.003Pb_{OM} - 0.116$	0.328	0.251	0.753
Fangzheng	$Pb_{rice} = 0.013Pb_{pH} 0.004Pb_{soil} - 0.004Pb_{OM} - 0.145$	0.419	0.006*	0.002*
Xiangshui	$Pb_{rice} = 0.01Pb_{pH} - 0.001Pb_{OM} - 0.039$	0.311	-0.076	0.608*
Jiansanjiang	$Pb_{rice} = 0.001Pb_{soil} - 0.007Pb_{pH} - 0.002Pb_{OM} + 0.040$	0.224	0.201	-0.416*

*Significant at the 0.05 level.

Study area	Element -	Single factor pollution index			
Study area		Average	Max	Min	
Chahayang	Pb	0.104	0.249	0.048	
Wuchang	Pb	0.151	0.470	0.068	
Fangzheng	Pb	0.085	0.537	0.014	
Xiangshui	Pb	0.117	0.236	0.058	
Jiansanjiang	Pb	0.120	0.275	0.059	

Table 7. Single factor pollution index and proportion of pollution grade in the study area.

Table 8. Single factor and comprehensive pollution index and comprehensive pollution grade in the area.

Study area	Single factor pollution index Pb	Comprehensive factor pollution index	Pollution index
Chahayang	0.104	0.135	No pollution
Wuchang	0.151	0.247	No pollution
Fangzheng	0.085	0.272	No pollution
Xiangshui	0.117	0.132	No pollution
Jiansanjiang	0.120	0.150	No pollution
Total	0.115	0.275	No pollution

not polluted by Pb, and the proportion of pollution-free in each region is 100%. From the evaluation results of the comprehensive pollution index (Table 8), the comprehensive pollution index of rice in the study area is 0.275, which is a level of pollution-free; the comprehensive pollution index of each region is sequentially ranked as Fangzheng> Wuchang> Jiansanjiang> Chahayang> Xiangshui, pollution The index is between 0.132 and 0.272. Due to the different geographical locations of different regions, and the level of economic development and the distribution of industrial structures also vary, the Pb content in rice varies from region to region. The results are similar to the results of non-carcinogenic risk assessment in Zhejiang(Zhao et al., 2009). According to the research results (Tables 7 and 8), the pollution levels of heavy metals in the study area are all pollution-free.

According to the results of the non-carcinogenic risk assessment (Table 9), the average intake of Pb (ADD) of adults and children is lower than the reference exposure dose (RfD), and the individual health risk index is less than 1, indicating that the daily exposure of Pb As for the amount of Pb, Pb has not caused any health risks to the human body. For each region, the health risks of adults and children caused by rice intake are ranked as Wuchang> Chahayang> Jiansanjiang> Fangzheng> Xianshui. Comparing the results of individual rice pollution evaluation in the study area, the pollution index in the Xiangshui area of rice in each region was the smallest, and the area was least affected by Pb pollution. Lead is an accumulative harmful element, which has relatively obvious toxic effects on the human nervous system and reproductive system, and is a heavy metal element with a high risk of carcinogenesis.

Due to the international nature of this experimental index parameter and the regionality of the study area, as well as the

 Table 9. Heavy metal Pb intake and non-carcinogenic risk in rice pathway.

Crowd	ADD		HQ	
Clowd	Adult	Child	Adult	Child
Chahayang	5.724×10-5	8.865×10-5	0.004	0.006
Wuchang	9.943×10-5	1.539×10^{-4}	0.007	0.010
Fangzheng	4.181×10 ⁻⁵	6.475×10 ⁻⁵	0.002	0.004
Xiangshui	5.644×10-5	8.741×10 ⁻⁵	0.001	0.006
Jiansanjiang	4.806×10 ⁻⁵	7.444×10 ⁻⁵	0.003	0.005

differences in the quality of the human body in different living environments, the results of this study have certain limitations and one-sidedness. In addition, since the impact of rice varieties was not considered in the research process of this article, in the subsequent research, the migration in the "soil-plant-human" system should be established on the basis of comprehensive consideration of various current impact factors Experimental research on transformation, transformation and bioavailability, with a view to providing data reference for agricultural land soil protection and food security.

4 Conclusion

This paper objectively evaluated the lead content of rice in five major production areas of Heilongjiang Province, which has certain reference value and practical significance. The research results show that the lead content in rice and soil in the study area exceeds the limit of China's national food hygiene standard, which is also determined by the different economic industries in different regions and the situation of cultivated land and other factors. The average lead content of brown rice in the study area was Chahayang 0.02 mg/kg, Wuchang 0.03 mg/kg, Fangzheng 0.017 mg/kg, Xiangshui 0.023 mg/kg and Jiansanjiang 0.024 mg/ kg, the lead content in the five regions did not exceed the lead content stipulated in China's national food hygiene standards, and the differences in lead content in brown rice, polished rice, and soil in the five regions studied were inconsistent. The prediction model developed in this study, including total soil heavy metals and pH, can significantly describe the transfer of lead in the soil-rice system of Chahayang, Fangzheng, Xiangshui, and Jiansanjiang paddy fields. In the meantime, the Wuchang area model remains to be studied.

The pollution index of the entire study area is less than 1. The comprehensive pollution index is 0.275> 1, which belongs to the non-pollution category; the comprehensive pollution index of each region is sequentially ranked as Fangzheng> Wuchang> Jiansanjiang> Chahayang> Xiangshui. The comprehensive pollution index is between 0.132 and 0.272, which is non-Category of pollution. The average daily intake (ADD) of lead for adults and children is lower than the reference dose (RfD), and the health risk index of heavy metal Pb for adults and children is lower than the maximum acceptable risk level recommended by USEPA, which will not cause health risk to human body.

Acknowledgements

This work was supported by the National Key Research and Development Program "2021YFD2100903", the National Key Research and Development Program (Rice and wheat moderate processing and product value-added key technology research and development and industrialization demonstration), the Department of Education, Heilongjiang Province (grant number [2018] No. 4), the cooperation research and application demonstration of refifined processing key technology of coarse cereals food (2018YFE0206300).

References

- Baker, A., McGrath, S. P., Sidoli, C., & Reeves, R. D. (1994). The possibility of in situ heavy metal decontamination of polluted soils using crops of metal-accumulating plants. *Resources, Conservation and Recycling*, 11(1-4), 41-49. http://dx.doi.org/10.1016/0921-3449(94)90077-9.
- Basta, N. T., Ryan, J. A., & Chaney, R. L. (2005). Trace element chemistry in residual-treated soil: key concepts and metal bioavailability. *Journal* of Environmental Quality, 34(1), 49-63. http://dx.doi.org/10.2134/ jeq2005.0049dup. PMid:15647534.
- Cai, L. M., Xu, Z. C., Qi, J. Y., Feng, Z. Z., & Xiang, T. S. (2015). Assessment of exposure to heavy metals and health risks among residents near Tonglushan mine in Hubei, China. *Chemosphere*, 127, 127-135. http://dx.doi.org/10.1016/j.chemosphere.2015.01.027. PMid:25676498.
- Cao, D. J., Shi, X. D., Li, H., Xie, P. P., Zhang, H. M., Deng, J. W., & Liang, Y. G. (2015). Effects of lead on tolerance, bioaccumulation, and antioxidative defense system of green algae, Cladophora. *Ecotoxicology and Environmental Safety*, 112, 231-237. http://dx.doi. org/10.1016/j.ecoenv.2014.11.007. PMid:25463875.
- Chang, C. Y., Yu, H. Y., Chen, J. J., Li, F. B., Zhang, H. H., & Liu, C. P. (2014). Accumulation of heavy metals in leaf vegetables from agricultural soils and associated potential health risks in the Pearl River Delta, South China. *Environmental Monitoring and Assessment*, 186(3), 1547-1560. http://dx.doi.org/10.1007/s10661-013-3472-0. PMid:24185814.
- Duan, X., Zhao, X., Wang, B., Chen, Y., & Cao, S. (2015). Highlights of the chinese exposure factors handbook. In X. Duan, X. Zhao, B.
 Wang, Y. Chen & S. Cao (Eds.), *Highlights of the chinese exposure factors handbook*. London: Academic Press.
- Dudka, S., Piotrowska, M., & Terelak, H. (1996). Transfer of cadmium, lead, and zinc from industrially contaminated soil to crop plants: a field study. *Environmental Pollution*, 94(2), 181-188. http://dx.doi. org/10.1016/S0269-7491(96)00069-3. PMid:15093504.
- Dutta, D., Mandal, B., & Mandal, L. N. (1989). Decrease in availability of zinc and copper in acidic to near neutral soils on submergence. *Soil Science*, 147(3), 187-195. http://dx.doi.org/10.1097/00010694-198903000-00005.
- Fang, Y., Sun, X., Yang, W., Ma, N., Xin, Z., Fu, J., Liu, X., Liu, M., Mariga, A. M., Zhu, X., & Hu, Q. (2014). Corrigendum to "Concentrations and health risks of lead, cadmium, arsenic, and mercury in rice and edible mushrooms in China". *Food Chemistry*, 151, 379. http:// dx.doi.org/10.1016/j.foodchem.2013.11.078.
- Giri, S., & Singh, A. K. (2015). Human health risk assessment via drinking water pathway due to metal contamination in the groundwater of Subarnarekha River Basin, India. *Environmental Monitoring and Assessment*, 187(3), 63. http://dx.doi.org/10.1007/s10661-015-4265-4. PMid:25647791.
- Haldar, M., & Mandal, L. N. (1979). Influence of soil moisture regimes and organic matter application on the extractable zn and cu content in rice soils. *Plant and Soil*, 53(1-2), 203-213. http://dx.doi. org/10.1007/BF02181891.

- Haldar, M., & Mandal, L. N. (1981). Effect of phosphorus and zinc on the growth and phosphorus, zinc, copper, iron and manganese nutrition of rice. *Plant and Soil*, 59(3), 415-425. http://dx.doi. org/10.1007/BF02184546.
- Jalali, M., & Meyari, A. (2022). Heavy metal contents, soil-to-plant transfer factors, and associated health risks in vegetables grown in western Iran. *Journal of Food Composition and Analysis*, 106, 104316. http://dx.doi.org/10.1016/j.jfca.2021.104316.
- Jung, M. C., & Thornton, I. (1997). Environmental contamination and seasonal variation of metals in soils, plants and waters in the paddy fields around a pb-zn mine in Korea. *The Science of the Total Environment*, 198(2), 105-121. http://dx.doi.org/10.1016/S0048-9697(97)05434-X. PMid:9167264.
- Kashem, M. A., & Singh, B. R. (2001). Metal availability in contaminated soils: I. Effects of flflooding and organic matter on changes in Eh, pH and solubility of Cd, Ni and Zn. *Nutrient Cycling in Agroecosystems*, 61(3), 247-255. http://dx.doi.org/10.1023/A:1013762204510.
- Lee, K.-J., Feng, Y. Y., Choi, D.-H., & Lee, B.-W. (2016). Lead accumulation and distribution in different rice cultivars. *Journal of Crop Science and Biotechnology*, 19(4), 323-328. http://dx.doi.org/10.1007/ s12892-016-0085-5.
- Li, N., Kang, Y., Pan, W., Zeng, L., Zhang, Q., & Luo, J. (2015). Concentration and transportation of heavy metals in vegetables and risk assessment of human exposure to bioaccessible heavy metals in soil near a waste-incinerator site, South China. *The Science of the Total Environment*, 521-522, 144-151. http://dx.doi.org/10.1016/j. scitotenv.2015.03.081. PMid:25829292.
- Liu, X., Gu, S., Yang, S., Deng, J., & Xu, J. (2021). Heavy metals in soilvegetable system around E-waste site and the health risk assessment. *The Science of the Total Environment*, 779, 146438. http://dx.doi. org/10.1016/j.scitotenv.2021.146438. PMid:33744561.
- Lu, Q., Xiao, Q., Guo, Y., Wang, Y., Cai, L., You, W., Zheng, X., & Lin, R. (2022). Pollution monitoring, risk assessment and target remediation of heavy metals in rice from a five-year investigation in Western Fujian region, China. *Journal of Hazardous Materials*, 424(Pt C), 127551. http://dx.doi.org/10.1016/j.jhazmat.2021.127551. PMid:34736193.
- Luo, H., Wang, Q., Guan, Q., Ma, Y., Ni, F., Yang, E., & Zhang, J. (2022). Heavy metal pollution levels, source apportionment and risk assessment in dust storms in key cities in Northwest China. *Journal* of Hazardous Materials, 422, 126878. http://dx.doi.org/10.1016/j. jhazmat.2021.126878. PMid:34418825.
- McBride, M. B. (2002). Cadmium uptake by crops estimated from soil total cd and ph. *Soil Science*, 167(1), 62-67. http://dx.doi. org/10.1097/00010694-200201000-00006.
- McLaughlin, M., Tiller, K., & Smart, M. (1996). Speciation of cadmium in soil solutions of saline/sodic soils and relationship with cadmium concentrations in potato tubers (*Solanum tuberosum* L.). *Soil Research*, 35(1), 1101-1102. http://dx.doi.org/10.1071/S96032.
- Means, B. (1989). *Risk-assessment guidance for Superfund*. *Volume 1. Human Health Evaluation Manual. Part A. Interim report (Final).* Washington: U.S. Environmental Protection Agency.
- Mehr, M. R., Keshavarzi, B., Moore, F., Sacchi, E., Lahijanzadeh, A. R., Eydivand, S., Jaafarzadeh, N., Naserian, S., Setti, M., & Rostami, S. (2016). Contamination level and human health hazard assessment of heavy metals and polycyclic aromatic hydrocarbons (pahs) in street dust deposited in mahshahr, southwest of iran. *Human and Ecological Risk Assessment*, 22(8), 1726-1748. http://dx.doi.org/10. 1080/10807039.2016.1219221.
- Mushak, P. (2003). Lead remediation and changes in human lead exposure: some physiological and biokinetic dimensions. *The Science*

of the Total Environment, 303(1-2), 35-50. http://dx.doi.org/10.1016/ S0048-9697(02)00358-3. PMid:12568763.

- Pan, L., Li, G., Li, J., Gao, J., Liu, Q., & Shi, B. (2022). Heavy metal enrichment in drinking water pipe scales and speciation change with water parameters. *The Science of the Total Environment*, 806(Pt 2), 150549. http://dx.doi.org/10.1016/j.scitotenv.2021.150549. PMid:34600211.
- Reddy, C. N., & Patrick, W. H. Jr. (1977). Effect of redox potential and ph on the uptake of cadmium and lead by rice plants. *Journal of Environmental Quality*, 6(3), 259-262. http://dx.doi.org/10.2134/ jeq1977.00472425000600030005x.
- Sajwan, K. S., & Lindsay, W. L. (1986). Effects of redox on zinc deficiency in paddy rice. Soil Science Society of America Journal, 50(5), 1264-1269. http://dx.doi.org/10.2136/sssaj1986.0361599500500050036x.
- Salam, A. K., Rizki, D. O., Santa, I., Supriatin, S., Septiana, L. M., Sarno, S., & Niswati, A. (2022). The biochar-improved growth-characteristics of corn (*Zea mays* L.) in a 22-years old heavy-metal contaminated tropical soil. *IOP Conference Series. Earth and Environmental Science*, 1034(1), 012045. http://dx.doi.org/10.1088/1755-1315/1034/1/012045.
- Silbergeld, E. K., Waalkes, M., & Rice, J. M. (2000). Lead as a carcinogen: experimental evidence and mechanisms of action. *American Journal* of Industrial Medicine, 38(3), 316-323. http://dx.doi.org/10.1002/1097-0274(200009)38:3<316::AID-AJIM11>3.0.CO;2-P. PMid:10940970.
- Simon, D. L., Maynard, E. J., & Thomas, K. D. (2007). Living in a sea of lead--changes in blood- and hand-lead of infants living near a smelter. *Journal of Exposure Science & Environmental Epidemiology*, 17(3), 248-259. http://dx.doi.org/10.1038/sj.jes.7500512. PMid:16823398.
- Singh, R. P., Tripathi, R. D., Dabas, S., Rizvi, S. M. H., Ali, M. B., Sinha, S. K., Gupta, D. K., Mishra, S., & Rai, U. N. (2003). Effect of lead on growth and nitrate assimilation of *Vigna radiata* (L.) Wilczek seedlings in a salt affected environment. *Chemosphere*, 52(7), 1245-1250. http://dx.doi.org/10.1016/S0045-6535(03)00318-7. PMid:12821005.
- Srivastava, R. K., Pandey, P., Rajpoot, R., Rani, A., & Dubey, R. S. (2014). Cadmium and lead interactive effects on oxidative stress and antioxidative responses in rice seedlings. *Protoplasma*, 251(5), 1047-1065. http://dx.doi.org/10.1007/s00709-014-0614-3. PMid:24482190.
- U.S. Environmental Protection Agency US EPA, Office of Emergency and Remedial Response. (1986). *Superfund public health evaluation manual*. Washington: US EPA.
- Wang, L. F., Bai, Y. X., & Gai, S. N. (2011). Single-factor and nemerow multi-factor index to assess heavy metals contamination in soils on railway side of Harbin-Suifenhe Railway in Northeastern China.

Applied Mechanics and Materials, 71(78), 3033-3036. http://dx.doi. org/10.4028/www.scientific.net/AMM.71-78.3033.

- Wang, Z., Yu, X., Geng, M., Wang, Z., Wang, Q., & Zeng, X. (2017). Accumulation of heavy metal in scalp hair of people exposed in Beijing sewage discharge channel sewage irrigation area in Tianjin, China. *Environmental Science and Pollution Research International*, 24(15), 13741-13748. http://dx.doi.org/10.1007/s11356-017-8884-x. PMid:28401388.
- Wei, B., & Yang, L. (2010). A review of heavy metal contaminations in urban soils, urban road dusts and agricultural soils from China. *Microchemical Journal*, 94(2), 99-107. http://dx.doi.org/10.1016/j. microc.2009.09.014.
- Xia, X., Yang, Z., Cui, Y., Li, Y., Hou, Q., & Yu, T. (2014). Soil heavy metal concentrations and their typical input and output fluxes on the southern song-nen plain, Heilongjiang Province, China. *Journal of Geochemical Exploration*, 139, 85-96. http://dx.doi.org/10.1016/j.gexplo.2013.06.008.
- Yan, T., Zhao, W., Yu, X., Li, H., Gao, Z., Ding, M., & Yue, J. (2022). Evaluating heavy metal pollution and potential risk of soil around a coal mining region of Tai'an City, China. *Alexandria Engineering Journal*, 61(3), 2156-2165. http://dx.doi.org/10.1016/j.aej.2021.08.013.
- Yu, R., Wang, Y., Wang, C., Yu, Y., Cui, Z., & Liu, J. (2017). Health risk assessment of heavy metals in soils and maize (*Zea mays* L.) from yushu, northeast china. *Human and Ecological Risk Assessment*, 23(6), 1493-1504. http://dx.doi.org/10.1080/10807039.2017.1327800.
- Zeng, F., Mao, Y., Cheng, W., Wu, F., & Zhang, G. (2008). Genotypic and environmental variation in chromium, cadmium and lead concentrations in rice. *Environmental Pollution*, 153(2), 309-314. http://dx.doi.org/10.1016/j.envpol.2007.08.022. PMid:17905495.
- Zeng, X., Wang, Z., Wang, J., Guo, J., Chen, X., & Zhuang, J. (2015). Health risk assessment of heavy metals via dietary intake of wheat grown in Tianjin sewage irrigation area. *Ecotoxicology (London, England)*, 24(10), 2115-2124. http://dx.doi.org/10.1007/s10646-015-1547-0. PMid:26433741.
- Zhang, G., Song, K., Huang, Q., Zhu, X., Gong, H., Ma, J., & Xu, H. (2022). Heavy metal pollution and net greenhouse gas emissions in a rice-wheat rotation system as influenced by partial organic substitution. *Journal of Environmental Management*, 307, 114599. http://dx.doi.org/10.1016/j.jenvman.2022.114599. PMid:35092887.
- Zhao, K., Zhang, W., Zhou, L., Liu, X., Xu, J., & Huang, P. (2009). Modeling transfer of heavy metals in soil-rice system and their risk assessment in paddy fields. *Environmental Earth Sciences*, 59(3), 519-527. http://dx.doi.org/10.1007/s12665-009-0049-x.