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A New Approach to Differentiate the Causes of Excessive Cadmium in Rice: Soil Cadmium Extractability or Rice Variety

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Abstract: In order to effectively decrease cadmium (Cd) in rice grains in contaminated paddy soil and maintain the safe production of rice, identifying excessive Cd in rice caused by rice varieties or soil Cd is critical, but it is currently lacking. In the present study, the soil ethylenediaminetetraacetic acid (EDTA)-extractable Cd (EDTA-Cd) and the bioaccumulation factors of rice based on EDTA-Cd ($BCF_{EDTA\text{-}Cd}$) were used to develop an approach to identify excessive Cd in rice caused by rice varieties or soil Cd. Based on an empirical soil–plant transfer model and species sensitivity distribution (SSD), $BCF_{EDTA\text{-}Cd}$ and EDTA-Cd were divided into five grades. The results showed that the five grades of the EDTA-Cd (minimum value less than 0.11 mg/kg and maximum value greater than 2.93 mg/kg) and $BCF_{EDTA\text{-}Cd}$ (minimum value less than 0.09 and maximum value greater than 1.40) were classified in the normal soil pH range. Further, the conversion equation between EDTA-Cd and diethylene triamine pentaacetic acid (DTPA)-Cd was obtained through linear regression analysis using 67 sets of soil data from the literature. In addition, the four selected rounding thresholds for the percentage of EDTA-Cd to total soil Cd (EDTA-Cd) (%) were 52.5, 67.5, 82.5, and 97.5%. A selected soil EDTA-Cd (%) (about 75%) can be used to identify the status of soil bioavailability, especially in soil with high background Cd. Finally, a set of 1084 pairs of rice and soil data for Cd-contaminated soils was used to investigate the respective contributions of rice varieties and soil Cd when Cd in rice exceeds the limit (0.2 mg/kg). Based on field experiment data, a systematic identification approach for the causes of rice Cd exceeding the limit, soil Cd or rice variety, was established and applied. In conclusion, under Cd exposure conditions, the importance of the causes of Cd in soil and rice varieties can be identified, and their contributions can be distinguished, thus helping to identify the causes of Cd contamination in rice.

Keywords: cadmium; rice; soil EDTA-Cd; bioaccumulation factor; contamination cause identification

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1. Introduction

With rapid industrialization and urbanization in China, 16% of soil samples and 19% of agricultural soils are contaminated mainly with heavy metals and metalloids, based on China's soil environmental quality limits and nationwide surveys. The soil cadmium (Cd) exceeds the standard for cultivated land in China at the highest rate, as high as 7.0%, among the heavy metals [1]. The average Cd concentration is 0.27 mg/kg in Chinese arable soil [2]. About 45.1% of the total soil Cd content in China exceeds the risk screening values for agricultural soil (GB 15618-2018) [3,4]. Furthermore, the highly toxic Cd is easily accumulated in soil and absorbed by crops, and its contamination has occurred, significantly exceeding the contamination grades of the other heavy metals [5]. The Chinese Ministry of Agriculture Rice Product Quality Inspection and Supervision Center noted that the Cd concentration in 10% of rice grains exceeded the upper limit (0.2 mg/kg) [6]. In China, a major source of human Cd intake is rice grains; the bioaccumulation factor (BCF) of rice (0.4) is higher than the reported Cd enrichment factors in wheat (0.25) and maize (0.16) [7]. Therefore, Cd contamination in rice, especially in paddy soil, is a crucial and serious issue to be settled [8]. For the agro-environmental sustainability of world rice production and food safety, effective remediation and control strategies are necessary to reduce Cd contamination in paddy soils [9]. Scientists from different fields have proposed different strategies for the management and remediation of Cd-contaminated paddy soils [10,11]. Although it is more critical to differentiate the causes of excessive Cd in rice from soil contamination or from rice varieties, this is currently lacking.

Reducing Cd accumulation in rice grains and controlling Cd transportation in rice is particularly important for the normal growth of crops and better human health [12]. Bioavailable Cd refers to the Cd in the soil that can be absorbed by rice, which is the main concern related to rice absorption and accumulation of Cd. Soil available Cd concentration serves as a precise indicator for evaluating the risk associated with Cd absorption and accumulation in rice [13]. The results have shown that the Cd content in the soil is the most significant factor influencing Cd accumulation in soil–rice systems; in particular, bioavailable Cd in soil contributed to 54.78% of the Cd in rice [7]. Compared with the total amount, the available amount of soil heavy metals obtained by the chemical extraction method can better analyze the law of contaminant migration, reflect the ecological risk, and predict contaminant accumulation between soil and crops [14]. Chelating agents (such as ethylenediaminetetraacetic acid (EDTA)) as a single extraction reagent comprise a common and effective method for determining the bioavailable state of soil metal elements. Some experiments have shown that the best chemical extract for heavy metal extraction is EDTA [15–17]. Previous studies have indicated that the EDTA-Cd could be used as an indicator to derive the threshold concentrations of Cd for rice food safety and better explain the accumulation of Cd in rice grains [18–20].

The bioavailability of Cd to rice mainly depends on the physical and chemical properties of the soil and the physiological and ecological characteristics of the rice. Among them, the physical and chemical properties that affect the bioavailability of Cd include soil redox potential, soil pH, organic matter content, etc. These factors affect the solubility of Cd and thus the absorption of Cd by rice [21]. Some studies have indicated that soil pH and soil organic matter (SOM) are significantly correlated with the accumulation of heavy metals in rice grains [22–25]. Strategies to improve soil physicochemical properties and fix Cd in contaminated soils are mainly some agronomic measures, including the application of soil amendments and fertilizers, and water and tillage management.

In addition, the uptake and transport of Cd may be significantly different among rice varieties. Bioaccumulation factors refer to the ratio of the experimentally measured Cd concentration in plants to the total concentration in soil, which has been used as an indicator to describe the characteristics of Cd transfer and to predict the concentrations of Cd in plant tissues [26]. Therefore, the bioaccumulation factors of rice based on the EDTA-Cd ($BCF_{EDTA\text{-}Cd}$) refer to the ratio of the experimentally measured Cd concentration in plants to the soil EDTA-extractable Cd (EDTA-Cd). Screening, breeding, and popularizing rice varieties with low Cd accumulation is one of the effective technical means to reduce the risk of Cd pollution in rice [27].

Bioaccumulation factor prediction models have been utilized by many countries and institutions to develop environmental quality standards for contaminants and to determine the grade of heavy metal contamination in soil. According to recent studies, from 2016 to 2020, the development of soil–crop transfer prediction models for the derivation of thresholds in contaminated farmland gradually became one of the research hotspots [28,29]. An empirical model was developed for setting soil Cd criteria [30]. Furthermore, recent research developed reliable soil–plant transfer and species sensitivity distribution (SSD) models that can be used to derive soil probabilistic criteria for Cd [31]. A study summarized five prediction models for Cd content in rice grains [32]. It was evident that pH, soil Cd, and SOM are major factors influencing Cd accumulation in rice grains. Two types of rice Cd prediction models (low-accumulation varieties and high-accumulation varieties) were established based on soil properties [33]. Previous studies have shown that by combining an empirical soil–plant transfer model with SSDs which were derived with the Burr type III distribution model, the bioaccumulation factor of Cd in rice grains could be divided into five grades [26].

The current approaches to manage and control excessive Cd in rice are mostly the arbitrary selection or combination of agricultural measures and rice varieties, and there is no systematic approach to separately identify the two factors affecting Cd content in rice and take targeted control. As previously stated, the aim of the present research was to differentiate the causes of excessive Cd in rice soil Cd extractability or rice cultivars for the safe production of rice. A specific approach was established to identify the contribution of the EDTA-Cd and $BCF_{EDTA\text{-}Cd}$ when rice Cd exceeds the limit. The overall steps are described briefly as follows: (1) develop the evaluation method of the EDTA-Cd and $BCF_{EDTA\text{-}Cd}$; (2) classify the $BCF_{EDTA\text{-}Cd}$, the EDTA-Cd (%), and the EDTA-Cd into the following categories: ultra-low risk, low risk, moderate risk, high risk, and ultra-high risk; and (3) apply the systematic identification method for the soil Cd extractability and rice variety causes when rice Cd exceeds the limit.

2. Materials and Methods

2.1. Data Collection

Data used in this research were collected from two sources. Firstly, 67 data sets were obtained from the published literature and the Chinese Core Journal databases (CNKI databases), Springer and ScienceDirect, by using the keywords "Cd", "DTPA", "EDTA", and "soil" (note: DTPA denotes diethylene triamine pentaacetic acid). The data were selected according to the following criteria: (1) soil was used as the medium (excluding hydroponics experiments), and (2) the total Cd concentration and pH value of the soil were obtained, and the total Cd concentration of the soil did not exceed 3 mg/kg in order to eliminate the soils contaminated with slag in mining areas. Then, 1084 data sets were collected from real data of rice fields in Hunan in 2016. The measured data for each sample include rice Cd concentration, soil total Cd concentration, EDTA-Cd concentration, DTPA-Cd concentration, and pH value. The data selection criterion was that the total Cd value in the soil should be greater than the EDTA-Cd value in the soil.

Among the collected data, the soil pH was measured using a pH meter and a water ratio of 1:2.5 (g/mL) [34]. Soil samples were dissolved with aqua regia. The rice was husked according to the Chinese Ministry of Agriculture's "The Testing Methods of Rice Qualities" (NY 147-1988) [35] and then digested with HNO_3-$HClO_4$ or HNO_3-H_2O_2 mixed solution according to the Chinese standard "Determination of Cadmium in Food" (GB 5009.15-2014) [36]. The available Cd concentrations in soil samples were extracted using DTPA and EDTA extraction solutions. The concentrations of total Cd in soil and Cd in rice grains were determined by flame atomic absorption spectrometry or by inductively coupled plasma mass spectrometry [37].

2.2. Data Analyses

2.2.1. Soil Factors

The EDTA-Cd (%) was calculated using a linear regression model established by using the relation between the total Cd and EDTA-Cd [38]. The EDTA-Cd (%) was calculated as follows:

$$EDTA\text{–}Cd\ (\%) = \frac{EDTA - Cd}{Soil - Cd} \times 100 \quad (1)$$

where EDTA-Cd (%) is the ratio of EDTA-Cd to total Cd, EDTA-Cd (mg/kg) is the concentration of soil EDTA-extractable Cd, and Soil-Cd (mg/kg) is the total Cd concentration in the soil.

In previous research, a conversion model for total Cd and EDTA-Cd concentrations in soil was obtained (coefficient of determination, R^2 = 0.77) [38]. In the present study, refitting data from the previous study while eliminating the values with large differences, Table 1 lists the descriptive summary statistics of the data on Cd concentration in soil and the EDTA-Cd (mg/kg) across the sample period. The concentration of the EDTA-Cd was calculated as previously described. Moreover, because of better accuracy, the average of the foregoing values was applied to the following equation:

$$EDTA\text{–}Cd = 0.578Soil\text{–}Cd\left(R^2 = 0.989,\ n = 36,\ p\ <\ 0.001\right) \quad (2)$$

where EDTA-Cd (mg/kg) is the concentration of soil EDTA-extractable Cd, n is the number of samples used to derive the equation, and p is the level of marginal significance within a statistical hypothesis test.

Table 1. Mean, standard error, maximum, and minimum for the EDTA-Cd (mg/kg).

Sample Size	**EDTA-Cd (mg/kg)**			
	Mean	**Standard Error**	**Maximum**	**Minimum**
36	0.4193	0.0271	0.7251	0.2286

2.2.2. Bioaccumulation Factors

Bioaccumulation factors (BCFs) were calculated as follows:

$$BCF = \frac{Grain - Cd}{Soil - Cd} \quad (3)$$

where Grain-Cd (mg/kg) is the Cd concentration of rice grains.

As a consequence, the bioaccumulation factors of rice based on the EDTA-Cd ($BCF_{EDTA\text{-}Cd}$) were computed as follows:

$$BCF_{EDTA-Cd} = \frac{Grain - Cd}{EDTA - Cd} \quad (4)$$

2.2.3. Transfer Factors

Transfer factors (TFs) were calculated as follows:

$$TF = \frac{Grain - Cd}{Straw - Cd} \quad (5)$$

where Straw-Cd (mg/kg) is the Cd concentration of straw.

2.2.4. Normalization of the BCF Data

The soil–plant transfer model used in the present study is as follows:

$$\log_{10} BCF_{total} = 0.943 - 0.205\ pH - 0.312\ \log(OC) \quad (6)$$

where BCF_{total} is the bioaccumulation factor, pH is the value of pH, OC (g/kg) is the soil organic carbon content, and 0.943 is the intrinsic sensitivity for evaluating the ability of rice to accumulate soil Cd [39]. The process of normalization for the $BCF_{EDTA\text{-}Cd}$ values was similar to that of the BCF_{total} values. The relation between the total Cd and EDTA-Cd was applied to convert the empirical soil–plant transfer model based on the total Cd concentration into the model with the EDTA-Cd concentration. Then, combining Equations (2) and (6), the empirical soil–plant transfer model based on EDTA-Cd is as follows [38]:

$$\log_{10} BCF_{EDTA-Cd} = 1.181 - 0.205pH - 0.312\log(OC) \quad (7)$$

2.2.5. SSD Curve Construction

Using probability distribution functions, the species sensitivity distribution (SSD) method can not only extrapolate the toxicological level between different species but also realize the risk assessment of pollutants at biological community and ecosystem levels. The SSD method is considered to be more accurate for the baseline values used to assess the quality of the soil environment. For modeling empirical SSD, a Burr type III distribution is used for data sets of 8 species or more [40]. The Burr type III model often presents the best fit SSD curves of toxicity data [41]. The parametric equation of the Burr type III function, F(x), is as follows:

$$F(x) = \frac{1}{\left[1 + \left(\frac{b}{x}\right)^{c}\right]^{k}} \tag{8}$$

$$HC(q) = \frac{b}{\left[\left(\frac{1}{q}\right)^{\frac{1}{k}} - 1\right]^{\frac{1}{c}}} \tag{9}$$

where $HC(q)$ is the soil concentration of Cd that can secure the food safety for 95% of the crop species in the SSD analysis when q is 0.05 mg/kg; x is a fitted parameter; and b, c, and k are the three parameters of the Burr type III distribution. As a product of Equation (9), HC_5 is the cumulative concentration corresponding to the proportion of hazardous species on the SSD curve when it reaches 5%; a value smaller than HC_5 means that it represents a more toxic heavy metal.

2.2.6. The Conversion of EDTA-Cd and DTPA-Cd

Among the 67 soil samples, the total Cd concentration in soil ranged from 0.18 to 2.98 mg/kg, the EDTA-Cd concentration in soil ranged from 0.063 to 1.678 mg/kg, and the DTPA-Cd concentration in soil ranged from 0.051 to 1.3 mg/kg. Therefore, through linear regression analysis, taking into account the variables (the total soil Cd and soil pH), the conversion equation (R^2 = 0.9707) of EDTA-Cd to DTPA-Cd was obtained as the follows:

$$EDTA\text{–}Cd = 1.3177(DTPA\text{–}Cd) + 0.01114pH + 0.04779(Soil\text{–}Cd) - 0.083 \tag{10}$$

where DTPA-Cd (mg/kg) is the concentration of soil DTPA-extractable Cd.

2.2.7. Analysis of Paddy Field Data

A set of data containing the pH value, grain Cd concentrations, the DTPA-Cd, and the total soil Cd at 1084 sampling points was surveyed on agricultural land contaminated with Cd from rice. According to Equation (10), the DTPA-Cd values were converted to the EDTA-Cd values, applying Equation (4) to calculate the $BCF_{EDTA\text{-}Cd}$ for each of these sampling points and applying Equation (1) to calculate the EDTA-Cd (%). Then, regarding data processing, the first step was to identify outliers, which refer to extreme values that are abnormally beyond the overall pattern of the variable distribution. Outliers can significantly affect the estimation process of statistical data (such as the mean and standard deviation of the sample), causing the values to be overestimated or underestimated [42]. Values outside the range of three standard deviations are usually marked as outliers [43]. Therefore, values outside the range of three standard deviations (SDs or σ) were eliminated (3σ = 3.71). After that, the number of samples was reduced to 1003, and the statistics in Table 2 were computed.

Table 2. Descriptive statistics on soil properties and Cd concentrations in soils and grains.

Variables		Minimum	Maximum	Mean	Median	SD
Soil–rice	Soil pH	4.52	7.86	5.56	5.44	0.51
(*n* = 1003)	Cd_{soil} (mg/kg)	0.105	1.195	0.368	0.333	0.163
	Cd_{EDTA} (mg/kg)	0.063	0.998	0.278	0.255	0.135
	Cd_{grain} (mg/kg)	0.011	2.096	0.429	0.379	0.257
	$BCF_{EDTA\text{-}Cd}$	0.057	3.694	1.660	1.569	0.811

Cd_{soil}, Cd_{EDTA}, and Cd_{grain} represent the Cd concentrations in soil, EDTA-extractable state, and grains, respectively; $BCF_{EDTA\text{-}Cd}$ represents the bioaccumulation factors of rice based on EDTA-Cd; SD denotes the standard deviation.

2.2.8. Determining the Respective Contributions of the Causes of Rice Cd Contamination

Comparison of the risk grade values of $BCF_{EDTA\text{-}Cd}$ with EDTA-Cd can trace the respective contributions of the different causes of Cd contamination in rice. The two most critical factors affecting Cd concentration in rice were obtained by comparison. The flow chart for the determination of the contributions of the causes is shown in Figure 1.

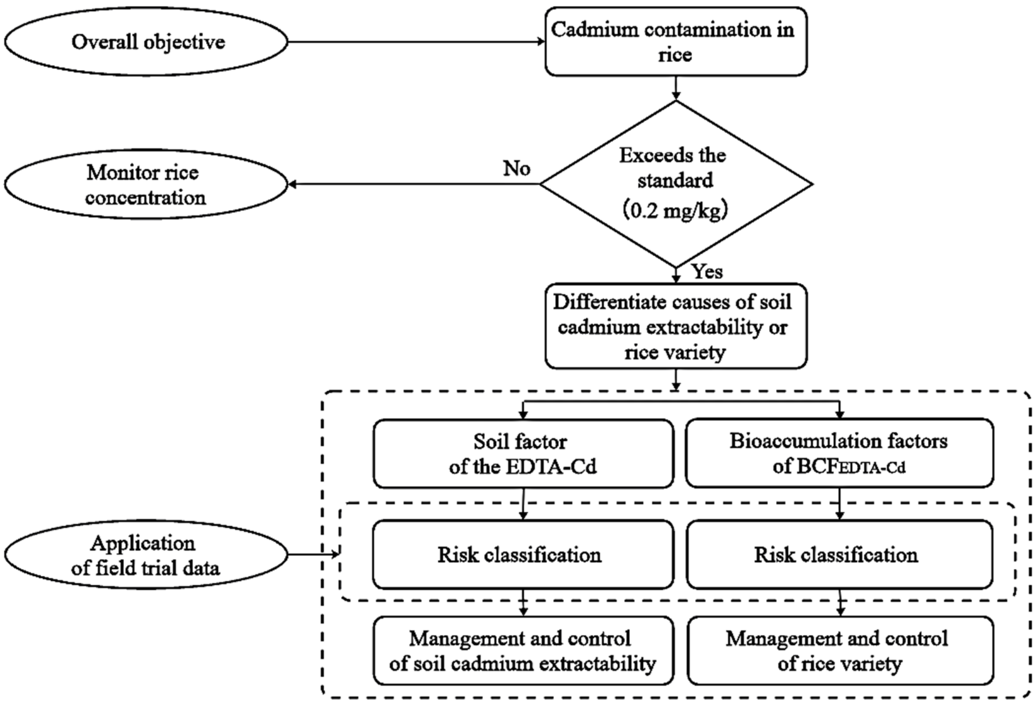

Figure 1. Flow chart for determining the respective contributions of the causes of excessive Cd in rice.

2.2.9. Statistics

The statistical package SPSS 26.0 was utilized for the statistical analysis and frequency distribution plot. Spreadsheets and figures were developed using Excel 2019. The difference at the level of $p < 0.05$ was considered significant.

3. Results

3.1. Soil Factor of the EDTA-Cd

3.1.1. Risk Classification of Soil-Cd

The causes of Cd contamination of rice were distinguished for the purpose of rice safety, so the risk classification of Cd in rice was the core concern, and the risk classification of the EDTA-Cd and $BCF_{EDTA\text{-}Cd}$ was conducted. Using the rice Cd contamination index as the evaluation standard, the rice Cd content limit value (0.2 mg/kg) was taken as the ultra-low risk value for the rice Cd content index, and two times the rice Cd content limit value was taken as the moderate risk critical value. Five risk grades of rice Cd and the corresponding rice content were obtained [44], as listed in Table 3.

Table 3. Risk classification of rice Cd.

Risk	Ultra-Low	Low	Moderate	High	Ultra-High
Rice Cd content (mg/kg)	<0.2	0.2–0.3	0.3–0.4	0.4–0.5	>0.5

Based on the Cd content of rice at each risk grade in Table 3, the total soil Cd concentration was calculated according to the SSD model (Equation (6)) based on soil properties (pH = 4, 5, 6, 7, and 8 and OC = 20). The total Cd values were divided into the five grades, from ultra-low to ultra-high risk. The derived ultra-low risk level critical value for total soil cadmium was slightly higher than the current soil quality standard, which was consistent with other research [45]. Table 4 lists the relation between the risk classes, soil pH, and Soil-Cd content.

Table 4. Risk classification of total Cd in soil.

Grade	Risk	Soil-Cd Content (mg/kg)				
		pH ≤ 4.5	4.5 < pH ≤ 5.5	5.5 < pH ≤ 6.5	6.5 < pH ≤ 7.5	pH > 7.5
1	Ultra-low	<0.19	<0.31	<0.49	<0.79	<1.26
2	Low	0.19–0.38	0.31–0.61	0.49–0.99	0.79–1.58	1.26–2.53
3	Moderate	0.38–0.58	0.61–0.92	0.99–1.48	1.58–2.38	2.53–3.81
4	High	0.58–0.77	0.92–1.23	1.48–1.97	2.38–3.17	3.81–5.07
5	Ultra-high	>0.77	>1.23	>1.97	>3.17	>5.07

Soil pH values were classified according to the soil quality standard of China (GB 15618-2018) [4]. The rounded thresholds at soil pH values of 4.0, 5.0, 6.0, 7.0, and 8.0 were used for scenarios of soil pH ≤ 4.5, 4.5–5.5, 5.5–6.5, 6.5–7.5, and >7.5, respectively. The scenario with soil OC was 20 g/kg.

3.1.2. Risk Classification of EDTA-Cd

According to Equation (7), the EDTA-Cd values were calculated based on soil properties (pH and OC). The EDTA-Cd values were divided into five risk grades, with a minimum value of less than 0.11 mg/kg and a maximum value greater than 2.93 mg/kg. Similar to the present results, other research found that the threshold value for Cd extractable by DTPA for safe rice production was 0.13 mg/kg when the pH in the soil was below 5.5 [20]. The risk classification of EDTA-Cd contamination is listed in Table 5.

Table 5. Risk classification of EDTA-Cd.

Grade	Risk	EDTA-Cd Content (mg/kg)				
		pH ≤ 4.5	4.5 < pH ≤ 5.5	5.5 < pH ≤ 6.5	6.5 < pH ≤ 7.5	pH > 7.5
1	Ultra-low	<0.11	<0.18	<0.29	<0.46	<0.73
2	Low	0.11–0.22	0.18–0.36	0.29–0.57	0.46–0.92	0.73–1.47
3	Moderate	0.22–0.33	0.36–0.53	0.57–0.86	0.92–1.37	1.47–2.20
4	High	0.33–0.44	0.53–0.71	0.86–1.14	1.37–1.83	2.20–2.93
5	Ultra-high	>0.44	>0.71	>1.14	>1.83	>2.93

Soil pH values were classified according to the soil quality standard of China (GB 15618-2018) [4]. The rounded thresholds at soil pH values of 4.0, 5.0, 6.0, 7.0, and 8.0 were used for scenarios of soil pH ≤ 4.5, 4.5–5.5, 5.5–6.5, 6.5–7.5, and >7.5, respectively. The scenario with soil OC was 20 g/kg.

3.1.3. Risk Classification of EDTA-Cd (%)

Regarding the EDTA-Cd (%), the statistics and frequency distribution table determined by frequency statistics are listed in Table 6. Figure 2 shows the cumulative distribution curve, normal distribution curve, and frequency histogram of the EDTA-Cd (%). Based on previous experience, the EDTA-Cd (%) ranged from 30 to 90% approximately. For convenience, the four selected rounding thresholds were 52.5, 67.5, 82.5, and 92.5%, corresponding to the 5th, 25th, 75th, and 95th percentile quantiles of the four cumulative frequency curves, respectively. The contamination risk grades were divided into five categories, in the following order: ultra-low risk was less than 52.5%, low risk was 52.5% to

67.5%, moderate risk was 67.5–82.5%, high risk was 82.5–92.5%, and ultra-high risk was greater than 92.5%. Thus, as shown in Figure 2, the median EDTA-Cd (%) of about 75% can be used to identify the status of soil bioavailability in practical applications, especially in soil with high background Cd. Other studies have found that the extraction rate of EDTA was the highest in acidic, neutral, and alkaline soils, up to 93.31% [15].

Table 6. Minimum (Min), maximum (Max), mean, and median of EDTA-Cd (%) and the percentile of the cumulative frequency curve.

EDTA-Cd (%)								
Sample Size	**Min (%)**	**Max (%)**	**Mean (%)**	**Median (%)**	**Percentile (%) Percentage (%)**			
1003	32	98	74	76	5	25	75	95
					51	67	83	93

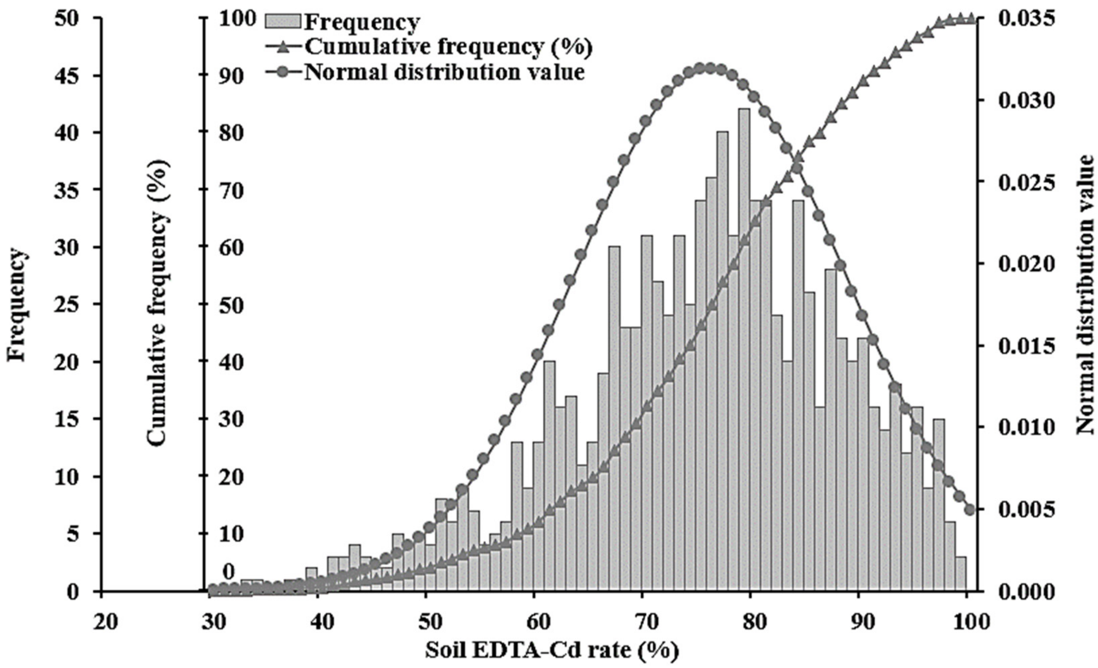

Figure 2. Cumulative distribution curve, normal distribution curve, and frequency histogram of the EDTA-Cd (%).

3.2. Bioaccumulation Factor of Rice Grains

Classification of Bioaccumulation Factors of Rice Based on EDTA-Cd

Referring to a previous study [26], combining the empirical soil–plant transfer model and species sensitivity distribution, the bioaccumulation factor of Cd in rice grains can be divided into five grades. Therefore, similarly, considering the relation between total soil Cd and EDTA-Cd, the BCF threshold under soil extractable Cd conditions was calculated. The normalized BCF decreased with increasing soil pH, and the critical value of BCF_{EDTA} at a pH greater than 6.5 was 47.8–62.8% lower than that at a pH less than or equal to 5.5. Under high-pH conditions, the variation range of BCF decreased. The risk classification of $BCF_{EDTA\text{-}Cd}$ is listed in Table 7.

Table 7. Bioaccumulation factors of rice based on EDTA-Cd ($BCF_{EDTA\text{-}Cd}$) under different risk grades.

Grade	Risk	pH ≤ 5.5	5.5 < pH ≤ 6.5	6.5 < pH ≤ 7.5	pH > 7.5
1	Ultra-low	<0.24	<0.17	<0.12	<0.09
2	Low	0.24–0.38	0.17–0.28	0.12–0.21	0.09–0.14
3	Moderate	0.38–0.57	0.28–0.42	0.21–0.29	0.14–0.21
4	High	0.57–1.40	0.42–1.00	0.29–0.73	0.21–0.52
5	Ultra-high	>1.40	>1.00	>0.73	>0.52

Soil pH values were classified according to the soil quality standard of China (GB 15618-2018) [4].

3.3. Comparison of Soil Factors and Extractable Bioaccumulation Factors at Measured Rice Concentrations

The risk grade values of EDTA-Cd and $BCF_{EDTA\text{-}Cd}$ for 1003 sample points are plotted in Figure 3. When the $BCF_{EDTA\text{-}Cd}$ grade value is higher than the EDTA-Cd grade value, the Cd content in rice grains is strongly affected by the extractable bioaccumulation factor of the rice grains. In this state, which occurred 97% of the time, selecting and cultivating low-Cd-accumulating rice cultivars might be a better option for the reduction in Cd accumulation in rice [9].

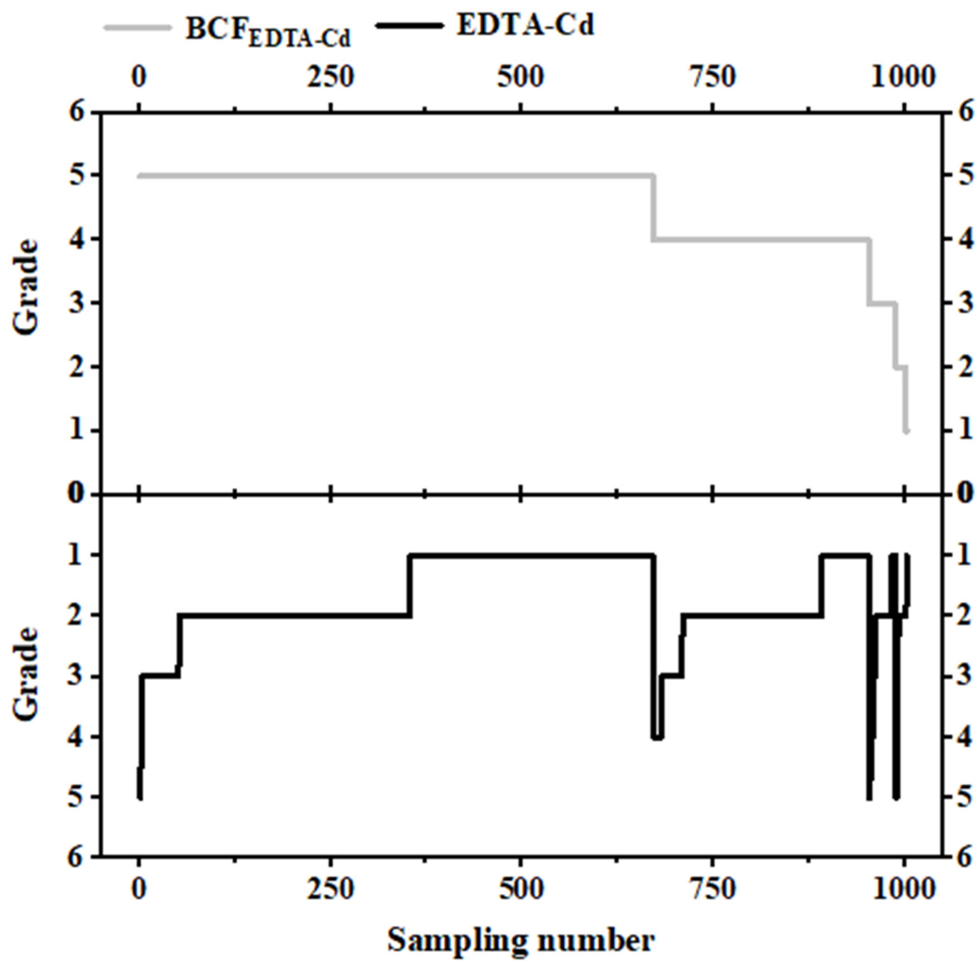

Figure 3. The Cd concentration of rice in all sample points and its corresponding risk grade of $BCF_{EDTA\text{-}Cd}$ and EDTA-Cd.

When the EDTA-Cd grade value exceeded the $BCF_{EDTA\text{-}Cd}$ grade value, which occurred 1% of the time, it suggests that decreasing the EDTA-extractable fractions of Cd is a more urgent method to control Cd contamination in rice. If the soil is acidic, increasing soil pH is the first priority. For example, the application of organic manure at a proper rate can reduce the accumulation and availability of Cd in the surface soil [46]. The addition of amendments of calcareous materials, such as lime, calcium carbonate, and calcium silicate, which can significantly raise soil pH and cause the immobilization of the labile or exchangeable Cd in soils, can help immobilize Cd^{2+} in the soils and reduce its availability [47,48]. A study indicated that the application of three different passivators could increase the soil pH by 0.62 to 0.86 units and reduce the Cd content in brown rice by 68% to 70% [49].

When the EDTA-Cd grade value and the $BCF_{EDTA\text{-}Cd}$ grade value were similar, which occurred 2% of the time, it meant that both rice varieties and soil properties were responsible for the excessive Cd in rice. Besides the foregoing approaches, irrigation management can also provide economically viable and environmentally friendly options for the remediation of Cd-contaminated soil. Water management also is a promising, controllable, and environmentally friendly approach to reduce Cd bioavailability and uptake [50]. A properly designed rice-based intercropping mode can provide an attractive alternative for Cd remediation in paddy fields [51]. Because the severity of Cd contamination in rice varies among different geographical locations, the primary causes and conditions of Cd contamination are different. For example, in northeast, central, southwest, and south China, the contents of Cd increase gradually from the northwest to southeast and from the northeast to southwest. In some provinces, mineral exploitation and industrial production are the main causes of Cd contamination, while in other provinces, these are irrigation with sewage and chemical fertilizer application [52]. Hence, it is necessary to adjust measures to better adapt to different conditions and choose the most effective and economical method to control excessive Cd in rice.

3.4. Applying Model by Using Experimental Field Data

The model for identifying cause contributions was applied to the experimental field data to test its feasibility and applicability. The data were derived from the previous literature. As shown in Table 8, in Scenario 1, the EDTA-Cd risk grade was low, and the $BCF_{EDTA\text{-}Cd}$ risk grade was ultra-low, while the risk of Cd contamination in rice was still ultra-low, so there was no need to take management measures. The only concern was to monitor the Cd content of rice. In Scenario 6, the Cd contamination risk grade of rice was ultra-high, so in theory, these fields should be fallow or should apply crop reform, but the respective contributions of the two factors, EDTA-Cd and $BCF_{EDTA\text{-}Cd}$, can still provide references for subsequent risk management and control.

In Scenario 4, the risk of Cd contamination in rice was high, and the contribution of soil Cd extractability was greater than that of rice variety in relation to excessive Cd content in rice. Hence, the cause of soil Cd extractability in the Cd contamination of rice was important. Research has demonstrated a significant inverse relation between the effective content of soil Cd and the pH value. Specifically, an increase in soil pH will lead to a reduction in the effective content of soil Cd, thereby decreasing its toxicity to rice [53]. Therefore, the risk grade of EDTA-Cd can be brought down by increasing the pH, especially in Scenario 4, where the pH value is 5.00. Research results also showed that adding zeolite increases the soil pH and the soil adsorption capacity for Cd, thereby significantly reducing soil Cd availability. Soil amendments such as lime are effective in immobilizing Cd in acidic soil but have limited effects in alkaline and neutral soil [54]. The Cd content in rice can also be effectively reduced by applying organic fertilizers and water management.

In Scenario 3, the risk grade of EDTA-Cd was lower than the risk grade of $BCF_{EDTA\text{-}Cd}$; thus, the cause of rice variety is of greater concern and had a greater contribution to Cd risk. The primary management of rice varieties can more quickly and specifically reduce the Cd content of rice, such as selecting low-Cd-accumulating rice cultivars. When the risk grade of rice variety factors is higher, rice with low-Cd-accumulating traits is more beneficial for reducing Cd concentration in rice grains.

Finally, it is worth mentioning that in Scenario 2, the risk grade of EDTA-Cd was equal to that of $BCF_{EDTA\text{-}Cd}$, which was moderate. Therefore, the two factors, EDTA-Cd and $BCF_{EDTA\text{-}Cd}$, had the same contribution to the excess of Cd in rice. In order to effectively reduce the Cd content of rice, the use of soil amendments and low-Cd rice varieties are both essential measures.

Table 8. Soil and rice data under different Cd contamination scenarios and corresponding management measures.

Scenario	[a] Cd_{grain} (mg/kg)	pH	[b] EDTA-Cd (mg/kg)	[c] $BCF_{EDTA\text{-}Cd}$	[d] Measures (Primacy)	Reference
1	0.04 Ultra-low	5.26	0.26 Low	Ultra-low		[55]
2	0.30 Moderate	5.60	0.77 Moderate	Moderate	A = B	[56]
3	0.38 Moderate	8.17	1.48 Moderate	High	A < B	[57]
4	0.40 High	5.00	0.86 Ultra-high	Moderate	A > B	[58]
5	0.81 Ultra-high	6.57	0.16 Ultra-low	Ultra-high		[59]

[a] Cd_{grain} is the Cd concentration of rice grains and the corresponding grade. [b] EDTA-Cd is the concentration of soil EDTA-extractable Cd and its corresponding grade. [c] $BCF_{EDTA\text{-}Cd}$ is the grade of bioaccumulation factors of rice based on EDTA-Cd. [d] Measures are management measures to reduce Cd content in rice, A is to add soil conditioners, B is to change the rice variety, and the primacy of the two measures is compared (>, =, <).

4. Discussion

4.1. Risk Assessment of EDTA-Cd in Soil

Different studies have different standards for the risk assessment of soil Cd contamination. Soil Cd contamination levels were classified as low, moderate, and high [60]. A recent study divided the Cd contamination status into four categories, which represent noncontaminated conditions and low, moderate, and high contamination grades [61]. The present research followed previous studies and classified soil Cd contamination into five grades, which are ultra-low risk, low risk, moderate risk, high risk, and ultra-high risk [44]. Some studies were focused on the risk assessment of total Cd in soil [62,63]. The threshold standard for EDTA-Cd was calculated to be 0.07 mg/kg when the pH value is less than 6.5 [18]. In the present study, the risk grade of EDTA-Cd contamination was divided into ultra-low, low, moderate, high, and ultra-high according to the degree of contamination from light to heavy. Meanwhile, the present research provides a reference for the Cd content standard under different extraction conditions and is conducive to the formulation of the standard of available Cd in the future.

4.2. Risk Assessment of $BCF_{EDTA\text{-}Cd}$

Soil risk assessment should consider not only soil characteristics but also differences between rice varieties [45]. Previous studies classified rice varieties into five grades based on the bioaccumulation of Cd in rice grains [26]. EDTA-Cd is more suitable to be used as a soil Cd threshold criterion for the food safety of rice crops [18]. If the extracted available Cd concentration is less than 0.19 mg/kg, 95% of the agricultural products grown in the soil could be below levels of regulatory concern [64]. Different from previous studies, the present research considered the importance of EDTA-Cd and proposed risk classification standards for $BCF_{EDTA\text{-}Cd}$. The evaluation of $BCF_{EDTA\text{-}Cd}$ can improve the accuracy of Cd bioaccumulation identification in rice grains.

4.3. Differentiating the Causes of Excessive Cadmium in Rice: Soil Cadmium Extractability or Rice Varieties

In the soil–rice system, the important factors affecting the absorption and accumulation of Cd by rice are the bioavailability of Cd in the soil and the capacity of Cd bioaccumulation in rice grains. The present study assessed the risk of soil Cd extractability and rice varieties. This was the first time that the two factors were distinguished, and the risk grade of the two factors can determine which factor needs to be the primary concern so as to more efficiently and accurately control Cd contamination in rice. However, the present research classifies the risk of Cd in the EDTA extraction state, and different Cd extraction methods can be considered in the future. The accuracy of this new approach still needs to be evaluated and improved.

5. Conclusions

In summary, unlike the total bioaccumulation factor and total soil Cd, the present research used the risk classification of soil Cd extractability and rice varieties to differentiate the causes of excessive Cd in rice. The EDTA-Cd, as well as the $BCF_{EDTA\text{-}Cd}$, could be divided into five grades. The present study derived a new approach which combines the EDTA-Cd with the $BCF_{EDTA\text{-}Cd}$ to identify the respective contributions of both causes in rice Cd contamination under Cd exposure. This work confirmed that the model can effectively demonstrate the contribution of the soil property factor and rice cultivar factor based on soil and rice data when rice Cd exceeds the standard. The difference between this method and the traditional method is that the two factors are analyzed separately. Then, by showing specific cases of Cd contamination in rice, it is recommended that managers primarily apply soil management or change rice varieties or both according to different situations. Overall, the present research provides support for the risk assessment of soil Cd contamination in rice production to ensure food safety. This present research contributes to appropriate land use management and accurate rice Cd contamination risk control in the future.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/agronomy14112519/s1.

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