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Effectiveness of Passivator Amendments and Optimized Fertilization for Ensuring the Food Safety of Rice and Wheat from Cadmium-Contaminated Farmland

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Abstract: The effect of various soil amendments on cadmium (Cd)-contaminated farmland was evaluated in terms of effectiveness, safety, economics, and simplicity. Experiments were conducted in May 2020, on Cd-contaminated land in Tong Ling, An Hui, China. The efficacy of optimized fertilization and heavy metal passivators—gypsum, bamboo charcoal, lime, and a compound passivator (mixture of bamboo charcoal, silicon fertilizer, gypsum, furfural residue, plant ash, and chicken manure)—was evaluated as amendments to reduce the uptake of Cd in rice. The results indicate that all treatments reduced the Cd bioavailability in Cd-contaminated soil and rice grain Cd accumulation to levels that meet national food safety standards. Moreover, the rice yield increased by 4.80 to 14.27% and the Cd content in rice grains decreased by 23.53 to 36.83%. The efficacy of Cd reduction in rice was as follows: optimized fertilization > lime > compound passivator > bamboo charcoal > gypsum powder. Wheat was planted after the rice season to test the effect of the soil amendment measures implemented during the rice season on crop growth in the next season. Wheat yield improved by 3.46 to 10.96%, and the grain Cd content decreased by 6.47 to 41.03%. The efficacy of Cd reduction in wheat was as follows: lime > compound passivator > optimized fertilization > gypsum powder > bamboo charcoal. Following the lime treatment, the wheat grain Cd content met national food safety standards. A comprehensive comparison was conducted to evaluate the safe utilization and economic effect of the passivator and optimized fertilization. The results reveal that optimized fertilization was the most effective treatment. The findings from this study provide a scientific basis for safe rice–wheat rotation systems on mildly Cd-contained farmland (0.3 mg/kg < Cd < 1.0 mg/kg) in the Yangtze River Basin.

Keywords: cadmium; passivator; rice; wheat; contaminant reduction; cadmium uptake

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1. Introduction

The large-scale use of chemical fertilizers, pesticides, and other chemical, along with the atmospheric deposition of heavy metals and industrial and mining waste discharge, have led to the large-scale heavy metal pollution of cultivated land in China [1–4]. It is reported that 19.4% of the cultivated land in China has excessive heavy metal levels, and Cd is the most severe [5,6]. Cadmium is a toxic heavy metal with strong bioavailability, and is easily taken up by plants [7,8]. Rice is one of the main food crops in China, and rice grains are prone to Cd accumulation, leading to Cd levels that exceed the allowed maximum concentration value (0.2 $mg \cdot kg^{-1}$) listed in the *China National Food Safety Standards for Pollutants in Food* (GB 2762-2017) [9]. Cadmium primarily enters rice through its root system as Cd^{2+}. In rice production regions with severe Cd contamination, the rice can become toxic when the Cd content accumulates to above 0.3 mg/kg, also resulting in a reduction in rice yield, grain carbohydrate, and amino acid levels [10–13]. Wheat is consumed by 60% of the world's population, and can absorb large amounts of Cd through its root system and transfer it to its shoots and grains, thereby posing a risk to human health [14]. Rice and wheat rotation is the main common crop planting pattern in the middle and lower reaches of the Yangtze River in China. It is estimated that the planting area of rice–wheat rotation accounts for approximately 16% of China's total rice production area, and the total rice–wheat rotation production accounts for more than 20% of China's grain output [15,16]. At present, the main management strategies for remediating Cd contamination in farmland include agronomic regulation, in-situ passivation, plant extraction and removal, soil leaching, and foliar resistance control [13,17–20]. Among them, in-situ passivation technology has been widely used due to its short remediation cycle and efficacy. In-situ passivation refers to the application of a passivator in polluted cultivated land, which reduces the bioavailability of heavy metals in the soil through ion exchange, adsorption, precipitation, and complexation with heavy metals, thereby reducing the absorption and enrichment of heavy metals in crops [21]. However, it is expensive and difficult to control, thereby reducing the passivation effect [22–26]. Optimized fertilization refers to optimizing the type and amount of chemical fertilizer applied, and improving the composition and ratio of fertilizer to safely utilize contaminated land, which has the benefits of being simple to manage and low in cost. Studies have shown that optimized fertilization can improve plant growth, impact the effective Cd content in soil, and reduce plant Cd uptake and accumulation [27–29]. In addition, the land used for rice–wheat rotation accounts for 10% of the total annual rice planting area [30]. Under hydroponic conditions, wheat absorbs much less Cd than rice [31]. The cultivation of soils previously passivated to reduce heavy metals with rice and wheat can reduce the amount of passivator needed to reduce the Cd content of wheat grains [32], improve the yield and nutrient content of rice and wheat grains [33,34], and maintain soil health [35,36]. The passivator needs to be used before the base fertilizer is applied. Compared with the use of a passivator, optimized fertilization is less costly and more convenient as it is only implemented once, when the base fertilizer is applied. However, there is a lack of comparative research on in-situ passivation and optimized fertilization. In this study, on-farm experiments were conducted in a rice–wheat rotation system. The impact of multiple passivators and optimized fertilization on crop (i.e., rice and wheat) Cd accumulation was evaluated and compared based on crop yield and other economic outcomes. The comparison between the two techniques provides an effective scientific basis for remediating Cd-contaminated farmland.

2. Materials and Methods

2.1. Experimental Materials

The experimental blocks were located in Cd-contaminated farmland in Tong Ling, An Hui, China, located in the middle and lower reaches of the Yangtze River. The soil used was conventional paddy soil. In the topsoil (depth ranging between 0 and 20 cm), the pH was 5.50, organic matter content was 22.77 g·kg^{-1}, total nitrogen (N) content was 1.06 g·kg^{-1}, effective phosphorus (P) content was 2.09 mg·kg^{-1}, available potassium (K) content was 270 mg·kg^{-1}, total Cd content was 1.601 mg·kg^{-1}, and effective Cd content was 0.581 mg·kg^{-1}. The total soil Cd content was 0.3–1.5 mg·kg^{-1}, which is above the soil contamination intervention value for agricultural land, as specified in China's *Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land*. Therefore, this land was classified as mildly to moderately Cd-contaminated soil. However, according to the classification of soil environmental quality in China, this field was classified as safe-to-use land [37]. The fertilizers and passivators used in this study were commercially available. Compound fertilizer (17-17-17) was purchased from Yang Feng Co. Ltd. (Jingmen, China). Urea was purchased from Liuguo Chemical Industry. Calcium magnesium phosphate and potassium sulfate were purchased from Hualv Ecology Environment Technology (Hefei, China). Lime was purchased from Tonglingshicheng Calcium Industry. Gypsum powder was purchased from LongCNY Gypsum ((Jingmen, China). The bamboo charcoal was purchased from Sanli New Fuel (Shangqiu, China). The compound passivator was purchased from Laimujia Biological Fertilizer (Bengbu, China) and the main components were furfural bamboo charcoal, silicon fertilizer, gypsum, slag, plant ash, and chicken manure.

2.2. Experimental Design

On-farm experiments were conducted with six treatment groups: blank control, lime, compound passivator, gypsum powder, bamboo charcoal, and optimized fertilization. The application rates of the experimental materials and fertilizer are presented in Table 1.

Table 1. Passivator and quantity.

Treatment	Material	Quantity Applied (kg·ha^{-1})	Properties
CK	N/A	/	/
L	Lime	2249	Alkaline cementitious material
CO	Compound passivator	2999	Alkaline inorganic and organic passivation materials
GP	Gypsum powder	1499	Alkaline crystalline powder
BC	Bamboo charcoal	2998	Hard texture, fine pores, and strong adsorption
OF	Calcium magnesium phosphate (OF)	560	Citron soluble soil improvement fertilizer

Note: CK, L, CO, GP and BC treatment groups were supplied with 450 kg·ha^{-1} compound fertilizer (17-17-17) as the base fertilizer during the rice season. A quantity of 150 kg·ha^{-1} of urea was applied as the tiller fertilizer, and 75 kg·ha^{-1} potassium sulfate was applied as the ear fertilizer. The OF treatment group was supplied with optimized fertilizer, i.e., 275 kg·ha^{-1} of compound fertilizer, 150 kg·ha^{-1} of potassium sulfate.

The experimental blocks were randomized. Each experimental block was triplicated. Therefore, there were 18 experimental blocks, each with an area of 20 m^2. Each block was separated by ridges covered with plastic films. All blocks were irrigated using clean water, therefore eliminating contamination. The rice variety was the locally grown Wan Jing Nuo No. 1 variety. The rice was transplanted, and the cultivation density was 30 × 13 cm.

In June 2020, compound fertilizer (17-17-17) was applied as a base fertilizer. Passivator and fertilizer were applied first into each block. The passivator was applied before applying the base fertilizer. When the passivator was added, the blocks were irrigated to a water-filled soil porosity (WFPS) above 80%. The soil was well-mixed using a rotary tiller. During the ploughing period, the water was not drained. Rice was planted seven days after ploughing, and top-dress tiller fertilizer was applied 20 days later. Additional fertilizer was applied during the rice booting stage. The yield of each block was measured separately after the rice matured, and soil and rice samples were also collected. In October 2020, when the rice was harvested, wheat was planted to test the effect of the soil amendment measures implemented during the rice season on crop growth in the next season. Hereafter, this effect is referred to as the aftereffect. The variety of wheat used was "Zhen Mai 15". Compound fertilizer (17-17-17) was used as a base fertilizer, and no passivator was used. Additional fertilizer was applied during the wheat booting stage.

2.3. Sample Collection

Soil samples were randomly collected at five locations with S-line sampling in each block when the rice or wheat matured. One kilogram of the topsoil (0–20 cm) was collected and indoor air-dried. The dried soil was then ground, thoroughly mixed, and retained for later experiments. The actual yield of the rice and wheat was measured at the mature stage. The rice and wheat samples were randomly collected from five plants once they had matured. Two parts of the samples were taken: grain and straw (leaves included in the straw sample). Plant samples were washed using ultrapure water and then placed into an oven at 105 °C for 30 min. The grain was dried at 70 °C. Brown rice samples were obtained by removing the bran, ground and thoroughly mixed, and passed through a 0.15 mm sieve for later use.

Economic data were calculated as follows.

The prices of rice, wheat, materials, manures, germchits, labor, and machinery were all derived from local market prices.

$$\text{Material cost (CNY·ha}^{-1}\text{)} = \text{Material unit price (CNY·kg}^{-1}\text{)} \times \text{Material applied (kg)}$$

$$\text{Manure cost (CNY·ha}^{-1}) = \text{Manure unit price (CNY·kg}^{-1}) \times \text{Manure applied (kg)}$$

$$\text{Agricultural total cost (CNY·ha}^{-1}) = \text{Material cost (CNY·ha}^{-1}) + \text{Manure cost (CNY·ha}^{-1}) + \text{Germchit cost (CNY·ha}^{-1}) + \text{Labor cost (CNY·ha}^{-1}) + \text{Machinery cost (CNY·ha}^{-1})$$

$$\text{Yield income (CNY·ha}^{-1}) = \text{Yield (kg·ha}^{-1}) \times \text{Local price (CNY·kg}^{-1})$$

$$\text{Increased net revenue (CNY·ha}^{-1}) = \text{Yield income (CNY·ha}^{-1}) - \text{Agricultural total cost (CNY·ha}^{-1})$$

2.4. Sample Analysis Method

The soil pH was determined using distilled water (soil–water ratio of 1:2.5) and measured using a pH meter (PHS-3E, Shanghai INESA Scientific Instrument Co., Ltd., Shanghai, China). The soil organic matter was oxidized with excess $K_2Cr_2O_7$ and H_2SO_4 using an oil bath, and the excess $K_2Cr_2O_7$ was titrated with $FeSO_4$ solution. The content of organic matter was calculated based on the difference in potassium dichromate consumption between the sample and blank. The soil total N was determined by the Kjeldahl distillation method [38]. The soil was boiled with concentrated sulfuric acid, so that all the N-containing compounds were converted into NH_4^+, which was distilled into a boric acid solution by a Kjeldahl nitrogen analyzer and then titrated with acid [38]. Soil available P was extracted using the $NaHCO_2$ solution, and the phosphorus in the extracted solution was measured by the molybdenum antimony anti-colorimetric method [39]. The available K in the soil was extracted by NH_4OAc and determined by a flame photometer [40]. The soil available Cd was extracted by a DTPA solution and measured by an atomic absorption spectrophotometer [41]. The Cd in the rice straw, rice grain, wheat straw and wheat grain was determined by nitric acid and hydrogen peroxide solution digestion and an atomic absorption spectrophotometer [42] (ZEEnit700 Atomic Absorption Spectrometer, Jena Analytical Instruments AG, Jena, Germany).

2.5. Statistical Data Analysis

Excel 2020 (Microsoft Corp., Redmond, WA, USA) and SPSS 26.0 (IBM Corp., Armonk, NY, USA) were used for statistical analysis. One-way analysis of variance (ANOVA) was used, and the difference between the treatment and control was analyzed using Duncan's method ($p < 0.05$ and $p < 0.01$). Origin 2022 (OriginLab Corp., Northampton, Massachusetts, USA) was used to visualize the results. Correlation analysis was conducted using the "corrplot" function and Person's correlation method.

3. Result and Analysis

3.1. The Effect of Passivator and Optimized Fertilization on Rice and Wheat Yield

The impacts of passivator and optimized fertilization on rice yield, and their after effects on wheat yield, are shown in Figure 1. The rice yield was 5892–6733 kg·ha^{-1}. The results indicate that compared with CK (blank control), the rice yields of all treatment groups were higher (4.8–14.27% higher). However, the increase was not statistically significant. The wheat yield was 5783–6417 kg·ha^{-1}. The treatments also increased wheat yield, by 3.46–10.96%, though this increase was not statistically significant.

3.2. The Effect of Passivator and Optimized Fertilization on Rice Cd Content

The effect of passivator and optimized fertilization on rice Cd content is shown in Figure 2a. The Cd content in rice straw ranged from 0.353 to 0.993 mg·kg^{-1}. Compared with CK (blank control), the treatment groups all showed a significantly lower rice straw Cd content. The reduction level was between 40.37% and 64.46%. The Cd reduction efficacy was shown to be as follows: OF (optimized fertilization) > L (lime) > CO (compound passivator) > GP (gypsum powder) > BC (bamboo charcoal).

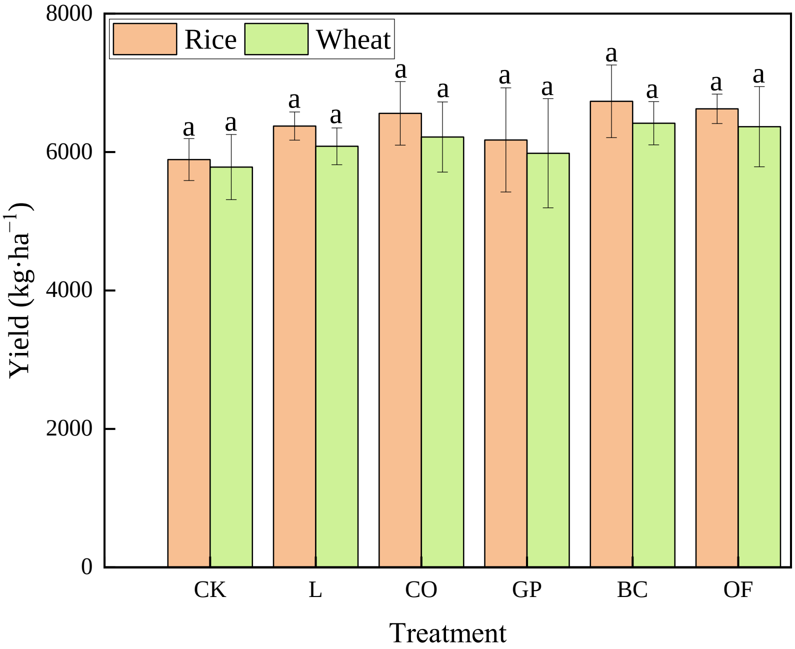

Figure 1. The effect of passivator and optimized fertilization on rice yield. Note: The value of each column represents the mean value of the three experimental blocks from a treatment. The error bar represents the value of the standard deviation. Columns marked with the same letters are not significantly different versus controls ($p > 0.05$), while the columns marked with different letters are significantly different compared with the control ($p < 0.05$). The meanings of the column, error bar, and the small letters in the figure below are consistent with the above explanation.

The rice grain Cd content ranged from 0.146 to 0.231 mg·kg^{-1}. Compared with CK (blank control), the Cd content in the treatment groups was reduced by 23.53% to 36.84%. The efficacy of grain Cd reduction ranked as: OF (optimized fertilization) > L (lime) > CO (compound passivator) > BC (bamboo charcoal) > GP (gypsum powder). The rice grain Cd content of the blank group was above the national standard control value (0.2 mg·kg^{-1}), while the rice grain Cd content of the treatment groups was below the control value [9]. Passivator and optimized fertilization treatments allowed the rice grains from mildly to moderately Cd-contaminated farmland to meet national standards.

3.3. The Effect of Passivator and Optimized Fertilization on Wheat Cd Content

The effect of passivator amendments and optimized fertilization on wheat Cd content is shown in Figure 2b. The Cd content of the wheat straw was 0.124 to 0.201 mg·kg^{-1}. Compared with CK (blank control), the wheat straw Cd content of the treatment groups was reduced by 6.63% to 38.44%. The Cd reduction of L (lime), CO (compound passivator), BC (gypsum powder), and OF (optimized fertilization) was significant ($p < 0.05$). The reduction ranked as: CO (compound passivator) > OF (optimized fertilization) > BC (gypsum powder) > L (lime) > OF (bamboo charcoal).

The wheat grain Cd content ranged from 0.092 to 0.156 mg·kg^{-1}. Compared with CK, the Cd content in the wheat grain was substantially reduced by 6.47% to 41.03%. The degree of reduction in L (lime), CO (compound passivator), GP (gypsum powder), and OF (optimized fertilization) was significant ($p < 0.05$). The degree of reduction ranked as: L (lime) > CO (compound passivator) > OF (optimized fertilization) > GP (gypsum powder) > BC (bamboo charcoal). The wheat grain Cd content with the after effects of L (lime) was below the national standard control value (0.1 mg·kg^{-1}), while that of the other treatment groups was above the control value [9].

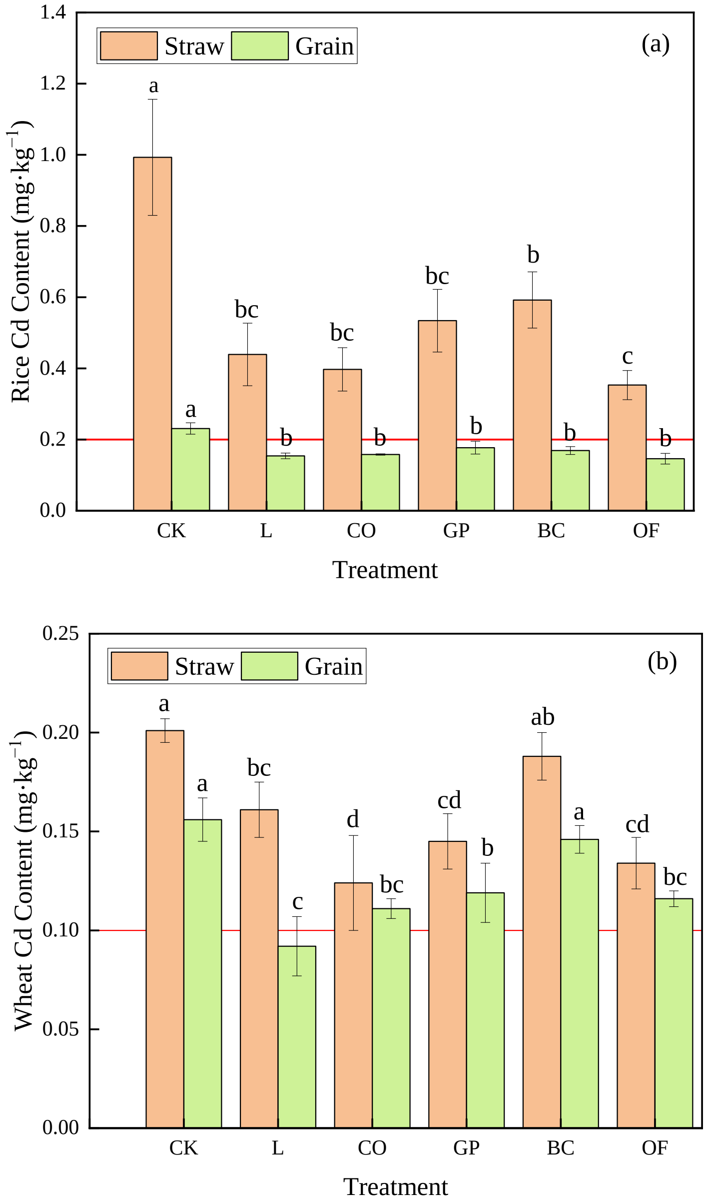

Figure 2. (**a**) The effect of passivator and optimized fertilization on rice cadmium content; (**b**) the effect of passivator and optimized fertilization on wheat cadmium content.

3.4. The Effect of Passivator and Optimized Fertilization on Soil Effective Cd (DTPA-Extracted Cd) Content after Rice and Wheat Harvest

The passivation and remediation effects of passivator amendment typically only reduce the effective Cd in the soil, rather than reducing the total Cd content. Therefore, the total effective soil Cd content was used as a quantitative parameter. The effect of passivator and optimized fertilization on the effective soil Cd content during the rice season is shown in Figure 3. The total effective soil Cd ranged from 0.182 to 0.502 mg·kg^{-1}. Compared with CK (blank control), the total effective Cd of all treatment groups was substantially reduced by 27.69% to 63/75%. The degree of reduction followed this pattern: OF (optimized fertilization) > CO (compound fertilizer) > GP (gypsum powder) > L (lime) > BC (bamboo charcoal).

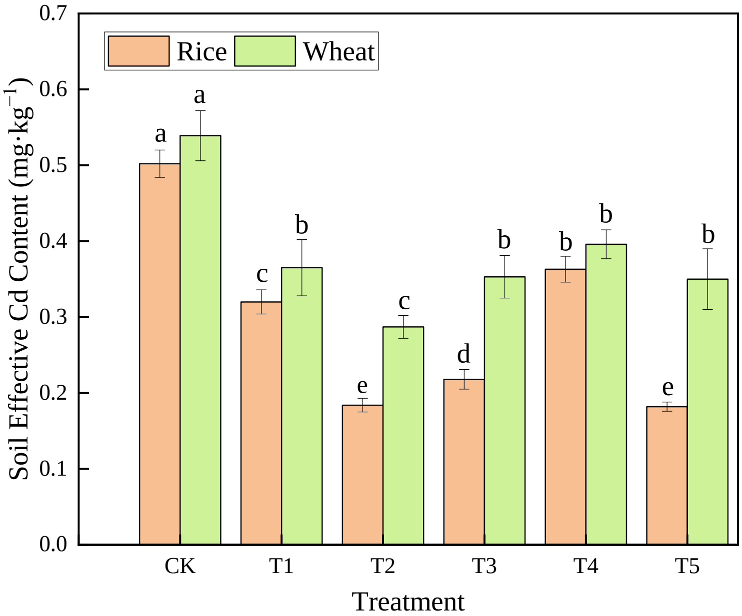

Figure 3. The effect of passivator and optimized fertilization on soil total effective cadmium.

The after effects of passivator amendments and optimized fertilization on the effective soil Cd content during the wheat season are shown in Figure 3. The total soil effective Cd content was about 0.287 to 0.539 mg·kg^{-1}. All treatment groups substantially reduced the total effective Cd content by 26.53% to 46.75%. The level of effective Cd reduction ranked as: CO (compound passivator) > OF (optimized fertilization) > GP (gypsum powder) > L (lime) > BC (bamboo charcoal).

3.5. The Effect of Passivator and Optimized Fertilization on the Bioaccumulation Factor of Rice and Wheat

The effect of passivator amendment and optimized fertilization on the rice and wheat Cd bioaccumulation factor is shown in Table 2. Compared with CK (blank control), the Cd bioaccumulation factor in the rice grains and shoots of each treatment group was significantly reduced by 23.91% to 37.15%, and 40.34% to 64.45%, respectively. These results indicate that all treatments inhibited Cd accumulation from the soil in the rice root system. Compared with CK (blank control), the after effect of each treatment group reduced the Cd bioaccumulation factor in the rice grain and stem by 6.04% to 40.76%, and by 6.22% to 38.17%, respectively. The after effects of L (lime), CO (compound passivator), GP (gypsum powder), and OF (optimized fertilization) were substantial ($p < 0.05$). The results indicate that the after effects of L (lime), CO (compound passivator), GP (gypsum powder), and OF (optimized fertilization) inhibited Cd accumulation from the soil to wheat.

3.6. Correlation between Rice Grain Cd Content, Rice Straw Cd Content, Rice Yield, and Soil Properties

The correlation analysis between rice grain Cd content, rice straw Cd content, rice yield, and soil properties is shown in Figure 4. The Cd contents in the rice grains have a strong positive correlation with the effective soil Cd content and rice straw Cd content (r = 0.74, r = 0.81). They also showed a strong negative correlation with soil pH (r = −0.66). The grain straw Cd content showed strong a positive correlation with soil effective Cd content (r = 0.82), and a strong negative correlation with soil pH (r = −0.58). The soil effective Cd content showed a strong negative correlation with soil pH (r = −0.54). The rice yield showed a strong negative correlation with rice grain Cd content (r = −0.55). The soil available phosphate content showed a strong positive correlation with soil pH (r = 0.75), and a strong negative correlation with the soil effective Cd content (r = −0.51). The soil total nitrogen had a strong positive correlation with organic matter content (r = 0.61).

Table 2. Effect of passivator and optimized fertilization on rice and wheat cadmium bioaccumulation factor.

Treatment	Rice		Wheat	
	BCF Grain/Soil	**BCF Stem/Soil**	**BCF Grain/Soil**	**BCF Stem/Soil**
CK	0.145 ± 0.010 a	0.620 ± 0.102 a	0.097 ± 0.007 a	0.125 ± 0.003 a
L	0.096 ± 0.005 b	0.274 ± 0.055 bc	0.057 ± 0.009 c	0.101 ± 0.007 bc
CO	0.099 ± 0.001 b	0.248 ± 0.038 bc	0.069 ± 0.003 bc	0.077 ± 0.012 d
GP	0.110 ± 0.012 b	0.334 ± 0.055 bc	0.074 ± 0.009 b	0.090 ± 0.007 cd
BC	0.105 ± 0.007 b	0.370 ± 0.049 b	0.091 ± 0.004 a	0.117 ± 0.006 ab
OF	0.091 ± 0.009 b	0.220 ± 0.026 c	0.073 ± 0.002 bc	0.084 ± 0.006 cd

Note: BCF is the enrichment coefficient of a plant organ. BCF = cadmium content in a plant organ/cadmium content in the soil. Mean values ± standard error; n = 3. Values marked with the same letters are not significantly different versus controls ($p > 0.05$), while the values marked with different letters are significantly different compared with the control ($p < 0.05$). The meaning of the small letters in the table below is consistent with the above explanation.

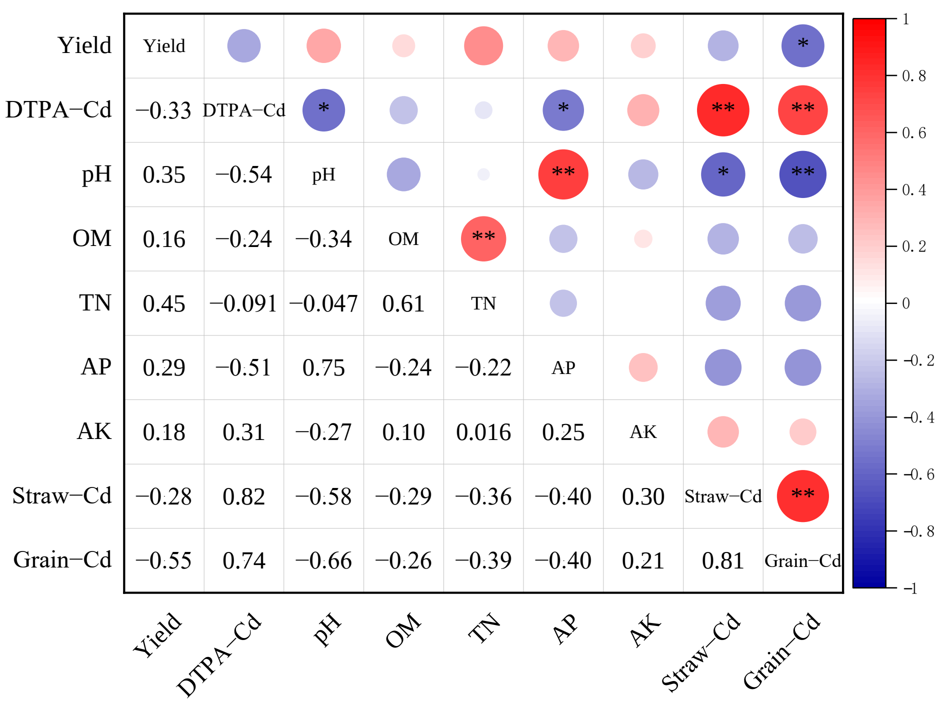

Figure 4. The correlation between rice grain/straw cadmium content, yield, and soil total effective cadmium content and physiochemical properties. Note: * $p \leq 0.05$, ** $p \leq 0.01$.

Research indicates that the application of lime, compound passivator, gypsum powder, bamboo charcoal, and optimized fertilization on Cd-contaminated farmland can somewhat increase soil pH. A change in soil pH could impact Cd conversion. A higher pH leads to a stronger absorption and fixation of Cd by soil colloids. As a result, the effective Cd content decreases [43]. Such results validate the strong positive correlation between soil pH and soil effective Cd content. Other research has indicated that the higher the Cd content in rice grains, the stronger the toxic effect on rice. Passivation can influence soil Cd and alleviate this negative impact, thereby improving rice yield [44]. The conclusions from past studies are consistent with the findings of this study.

3.7. Correlation between Wheat Grain Cd Content, Wheat Straw Cd Content, Rice Yield, and Soil Properties

The correlations between wheat grain Cd content, wheat straw Cd content, wheat yield, and soil properties are shown in Figure 5. The wheat grain Cd content showed a strong positive correlation with effective soil Cd content and wheat straw Cd content (r = 0.67, r = 0.67). It also showed a strong negative correlation with soil pH (r = −0.41). The wheat straw Cd content showed a strong positive correlation with soil effective Cd content (r = 0.81), and a negative correlation with soil pH (r = −0.85). The effective soil Cd content showed a strong negative correlation with soil pH (r = −0.73). The soil available potassium content showed a strong negative correlation with the wheat grain Cd content, the wheat straw Cd content, and the soil effective Cd content (r = −0.75, r = −0.47, and r = −0.58, respectively).

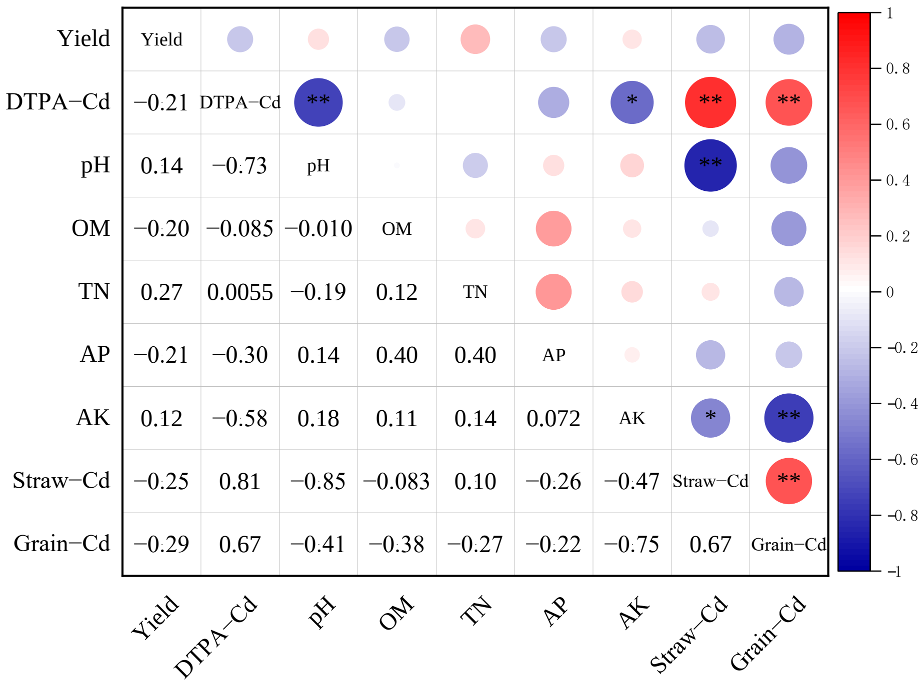

Figure 5. The correlation between wheat grain/straw cadmium content, yield, and soil effective cadmium content and other physiochemical properties. Note: * $p \leq 0.05$ ** $p \leq 0.01$.

During the second season, under the effect of passivator and optimized fertilization (wheat season), the effective soil Cd was still influenced by soil pH. The wheat yield showed a strong negative correlation with wheat grain Cd content. The results indicate that the wheat yield decreased with increasing grain Cd content, which is consistent with other studies. However, no solid conclusion exists regarding the correlation between wheat yield and grain Cd content [45–47].

3.8. The Effects of Passivator and Optimized Fertilization on Soil Physiochemical Properties

The effects of passivator amendment and optimized fertilization on rice soil physiochemical properties are shown in Table 3. Different passivators had a dramatically different impact on soil physiochemical properties. All parameters showed increasing trends, which indicated that the soil fertility was improved to some extent. Compared with CK (control group), the soil pH of the treatment groups increased by 0.03 to 0.18 units. The soil organic matter content after L (lime) and CO (compound passivator) increased by 42.21% and 84.72%, respectively. The soil total N content after L (lime), CO (compound passivator), and BC (bamboo charcoal) increased from 10.58% to 14.10%. The soil effective P content increased from 12.46% to 69.43% after CO (compound passivator), BC (bamboo charcoal), and OF (optimized fertilization). The available soil K increased by 1.53% to 11.68% after L (lime) and BC (bamboo charcoal).

Table 3. Effects of experiments on soil physiochemical properties.

Type	Treatment	pH	OM ($g{\cdot}kg^{-1}$)	TN ($mg{\cdot}kg^{-1}$)	AP ($mg{\cdot}kg^{-1}$)	AK ($mg{\cdot}kg^{-1}$)
Rice	CK	5.59 ± 0.05 d	19.61 ± 0.48 c	1.04 ± 0.03 b	11.96 ± 0.80 d	197 ± 4.71 b
	L	6.00 ± 0.11 b	28.20 ± 0.53 b	1.19 ± 0.07 a	10.28 ± 0.23 e	140 ± 7.76 d
	CO	5.78 ± 0.06 cd	36.22 ± 0.52 a	1.15 ± 0.02 a	15.10 ± 0.55 b	200 ± 8.16 b
	GP	5.80 ± 0.10 c	19.72 ± 1.41 c	1.06 ± 0.02 b	11.48 ± 0.64 de	147 ± 4.71 d
	BC	5.87 ± 0.04 bc	21.62 ± 1.25 c	1.15 ± 0.02 a	13.45 ± 0.50 c	220 ± 8.16 a
	OF	6.60 ± 0.09 a	16.09 ± 0.93 d	1.06 ± 0.02 b	20.26 ± 0.24 a	172 ± 2.36 c
Wheat	CK	5.16 ± 0.02 c	32.63 ± 1.64 ab	1.57 ± 0.07 a	8.89 ± 0.38 b	165 ± 4.08 c
	L	5.19 ± 0.01 c	35.48 ± 1.54 a	1.64 ± 0.01 a	10.37 ± 0.47 b	252 ± 6.24 a
	CO	5.42 ± 0.05 a	32.95 ± 1.95 a	1.51 ± 0.04 a	10.56 ± 0.87 b	203 ± 9.43 b
	GP	5.27 ± 0.04 b	35.07 ± 0.57 a	1.69 ± 0.14 a	17.29 ± 0.58 a	210 ± 6.53 b
	BC	5.19 ± 0.01 c	27.68 ± 1.38 c	1.59 ± 0.19 a	9.69 ± 1.38 b	200 ± 4.08 b
	OF	5.32 ± 0.02 b	28.65 ± 2.42 bc	1.61 ± 0.03 a	9.14 ± 0.52 b	240 ± 2.45 a

OM: organic matter; TN: total nitrogen; AP: available phosphate; AK: available potassium.

The effects of passivator and optimized fertilization on wheat soil physiochemical properties are shown in Table 3. The passivator after effects differed in the wheat soil. Compared with CK (blank control), the soil pH generally increased by 0.03 to 0.26 units. The after effect of BC (gypsum powder) increased the effective soil P by 94.49%, and the after effect of each treatment increased the available K by 21.21% to 52.53%.

3.9. Economics of the Application of Passivator and Optimized Fertilization

The Cd content of the rice grains from passivator and optimized fertilization treatments met the Chinese national food safety standards, while the CK rice grains did not. The economic impact of a passivator and optimized fertilization on rice is shown in Table 4. The agricultural cost was CNY 8250–17,912$\cdot ha^{-1}$ per season. Based on the local rice yield and commodity grain price, the net profit was CNY 321$\cdot ha^{-1}$. With safe production, the net profit could be CNY 5658–11,625$\cdot ha^{-1}$ (minus agricultural cost and assuming excessive grains meet the feed standard of Cd < 1 $mg{\cdot}kg^{-1}$ [48], a purchase price of CNY 1.5/kg, a qualified grain price of CNY 3.0/kg, and a grain yield of 5892–6733 $kg{\cdot}ha^{-1}$). The safe utilization of farmland can be realized through the application of a passivator, and as a result, the income of farmers can be increased.

The net profit of each treatment ranked as: OF (optimized fertilization) > L (lime) > CO (compound passivator) > GP (gypsum powder) > BC (bamboo charcoal) > CK (blank control). Overall, optimized fertilization had more considerable economic benefits, and is therefore more suitable.

Passivator and optimized fertilization were only used during the rice season. Except for L (lime), the after effects of the other treatments did not reduce the wheat grain Cd concentration to be within the national food safety standard (0.1 $mg{\cdot}kg^{-1}$) [9]. The economic after effect of passivator amendment and optimized fertilization is shown in Table 4. The agricultural cost per season was CNY 7223$\cdot ha^{-1}$. Based on the wheat yield and commodity grain price, assuming that the excessive wheat is sold as feed materials, the net profit can reach up to CNY 3197 to 4328$\cdot ha^{-1}$. Wheat that meets standards can be sold at normal commodity grain prices. Therefore, the net profit can be CNY 7377$\cdot ha^{-1}$ (minus agricultural costs and assuming that excessive grains meet the feed standard of Cd < 1 $mg{\cdot}kg^{-1}$) [46], with a purchase price of CNY 1.8/kg, a qualified grain price of CNY 2.4/kg, and a wheat yield of 5783 to 6417 $kg{\cdot}ha^{-1}$.

Table 4. Comparison of the economic effects between passivator and optimized fertilization.

Treatment		Agricultural Cost (CNY·ha^{-1})						Yield (kg·ha^{-1})	Yield Income (CNY·ha^{-1})	Increased Net Revenue (CNY·ha^{-1})
		Material	Manure	Germchit	Labor	Machinery	Total			
Rice	CK	/	3668	750	2600	1500	8518	5892	8838	321
	L	1799	3668	750	3000	1500	10,717	6375	19,125	8408
	CO	5098	3668	750	3000	1500	14,016	6558	19,674	5658
	GP	4497	3668	750	3000	1500	13,415	6175	18,525	5111
	BC	8994	3668	750	3000	1500	17,912	6733	20,199	2288
	OF	560	2840	750	2600	1500	8250	6625	19,875	11,625
Wheat	CK	/	3023	900	1800	1500	7223	5783	10,409	3187
	L	/	3023	900	1800	1500	7223	6083	14,599	7377
	CO	/	3023	900	1800	1500	7223	6217	11,191	3968
	GP	/	3023	900	1800	1500	7223	5983	10,769	3547
	BC	/	3023	900	1800	1500	7223	6417	11,551	4328
	OF	/	3023	900	1800	1500	7223	6367	11,461	4238
All year round	CK	0	6690	1650	4400	3000	15,740	/	19,247	3507
	L	1799	6690	1650	4800	3000	17,939	/	33,724	15,785
	CO	5098	6690	1650	4600	3000	21,038	/	30,865	9626
	GP	4497	6690	1650	4800	3000	20,637	/	29,294	8657
	BC	8994	6690	1650	4800	3000	25,134	/	31,750	6616
	OF	560	7153	1650	4600	3000	15,473	/	31,336	15,863

Labor fee including material application fee, transplanting fee, land loosening fee, and farmland management fee. The machine fee includes the rice harvesting fee.

The net profit of each treatment is ranked as follows: L (lime) > BC (bamboo charcoal) > OF (optimized fertilization) > CO (compound passivator) > GP (gypsum powder) > CK (blank control).

The economic effects of passivator amendment and optimized fertilization on crops (rice and wheat) are shown in Table 4. The agricultural cost per season was CNY 15,473 to 25,134·ha^{-1}. The net profit was CNY 3507 to 15,863·ha^{-1}. The net profit of each treatment ranked as: OF (optimized fertilization) > L (lime) > CO (compound passivator) > GP (gypsum powder) > BC (bamboo charcoal) > CK (blank control).

4. Discussion

4.1. The Effect of Passivator and Optimized Fertilization on the Safe Utilization of Rice

The results from this study indicate that rice grain Cd content ranked as: optimized fertilization > lime > compound passivator > bamboo charcoal > gypsum powder. Lime, compound passivator, bamboo charcoal, and gypsum powder can significantly reduce the Cd content in rice grains, and optimized fertilization has the best Cd reduction effect compared with other passivators. Zaniewicz-Bajkowska et al. [49] showed that the application of lime increased the soil pH, which led to the precipitation of active Cd. The compound passivator contained furfural residue, plant ash, and chicken manure. The furfural residue contained 45% cellulose and 45% lignin, which likely enhanced soil fertility [50]. The plant ash has a certain alkalinity, which introduces weak negatively charged groups such as hydroxyl and carboxyl. It can further increase the specific surface area and porosity, as well as the absorption ability [51]. Chicken manure increases the organic matter content and total soil N [52], which benefits the effective absorption and complexation of soil Cd. The bamboo charcoal effectively adsorbed the Cd in the water, attributed to its loose and porous structure [53]. Such a structure led to a large specific surface area, which facilitated the strong absorption capacity of Cd [54]. Through an indoor soil leaching experiment, Qin et al. [55] found that bamboo charcoal reduced the available Cd content in the soil by 40%. The gypsum powder was alkaline and rich in Cd and sulfate ions, which could form precipitates with Cd [56]. Compared with passivation, optimized fertilization led to improved food safety by reducing the rice grain Cd content by 36.84%. In this study, optimized fertilization was achieved by substituting monoammonium phosphate with calcium magnesium phosphate, and substituting potassium chloride with potassium sulfate. The calcium magnesium phosphate not only provided P, but also acted as a passivator. It could passivate heavy metals by forming insoluble P precipitates with them. Relevant research has shown that the application of calcium magnesium phosphate inhibits the absorption of Cd by plants, which is consistent with the results from this study. Yang et al. [57] found that the fertilizer composed of calcium carbonate, metakaolin, calcium, and magnesium phosphate could effectively reduce the Cd concentration in weak acid and salt extracts, and the rice grain Cd content was reduced by 74.1% when the compound fertilizer was applied at 1.6% (w/w). Zhao et al. [58] found that calcium magnesium phosphate inhibited the absorption of Cd by the plant root through the formation of iron plaque on the root surface. Cadmium absorption into the rice grain could be inhibited at a fertilizer amount between 0.5 and 2.5 g kg^{-1}. It could also facilitate the formation of free amino acids in the rice grain, thereby improving rice quality. Wang et al. [59] found through a 10-year farmland study that 1% (w/w) of calcium magnesium phosphate could reduce Cd accumulation in *Artemisia* by 40%. This is a stable and safe method for passivating heavy metals in soil, and is therefore suitable for the long-term passivation of Cd. Luo et al. [60] found that the rice grain Cd content was reduced by 56.14% when calcium magnesium phosphate was used at an amount of 37.5 $g{\cdot}m^{-2}$. Luo et al. [60] also found that the application of calcium–magnesia phosphate fertilizer promoted the transport of Cd from the root to the stalk to a certain extent, and inhibited the transport of Cd from the stalk to the grain, which was conducive to the accumulation of Cd in the rice straw. The same result was obtained herein, in that optimized fertilization reduced the enrichment coefficient of Cd in rice. This facilitated the accumulation of Cd in the rice straw. The type of K fertilizer affects the transport and accumulation of Cd in soil–crop systems. Research indicates that in Cd-contaminated land, potassium chloride is the least desirable fertilizer because it promotes the formation of Cd-chloride complexes, which can increase the mobility of Cd in the soil and exacerbate Cd accumulation in plants [61,62]. Chen et al. [63] found that the application of potassium sulfate reduced the phyto-availability of wheat on Cd, which is consistent with the effect of potassium chloride found in this study. The results from this study combined with earlier results indicate that the application of calcium magnesium phosphate and potassium sulfate can jointly reduce rice Cd accumulation from Cd-contaminated farmland. Comprehensive seasonal analysis indicates that compared with passivator treatment, optimized fertilization achieved a more desirable food safety outcome.

4.2. Comparison of the after Effects of Passivator Amendment and Optimized Fertilization on the Safe Utilization of Wheat

The results of this study show that compared with optimized fertilization and other passive agents, lime could reduce the bioavailability of soil Cd for a longer period of time, keep the soil available Cd content low, and allow the wheat grain Cd content to meet the national food safety standard (<0.1 $mg{\cdot}kg^{-1}$). Compared with the soil Cd content during the rice season, the soil content during the wheat season somewhat increased. This was attributed to the limited capacity and action time of the passivator. After two seasons, passivators may become saturated by Cd contaminants and other soil compounds [64]. So, the effect might not be greater with increased time. As time progresses, the fixed Cd might be released again [65]. Research has indicated that the action of lime, as a passivator, could be as long as a year and a half in acidic heavy metal-contaminated soil [66]. In this study, lime kept the grain Cd level in rice and wheat below the limit for one year. Therefore, lime has the optimal after effect. Other passivators such as compound passivator, gypsum powder, and bamboo charcoal did not maintain a long after effect, and should be reapplied after one season.

4.3. Comparison between the Economic and Environmental Effects of Passivator Amendment and Optimized Fertilization

This study demonstrates that passivator and optimized fertilization could substantially reduce the grain Cd level to below the National Food Safety Standards limit. No other treatment except for lime treatment could reduce the wheat grain Cd content below the limit. The economic benefit per hectare outweighed the safe utilization cost. Therefore, safe utilization could reduce the economic loss as a result of heavy metal contamination for local farmers. Optimized fertilization treatment led to the highest net profits, followed by lime. The profit difference was CNY 78·ha^{-1}. Over the long term, optimized fertilization had more environmental benefits than lime treatment. The long-term use of lime could lead to soil compaction and organic matter degradation, resulting in acidic soil [67–69]. The long-term use of lime can also reduce the effectivity of K and Mg in the soil [70]. Optimized fertilization also has economic and environmental benefits, as well as increasing crop yield in the long term [71,72]. The findings from this study suggest that optimized fertilization is the most suitable treatment for long-term implementation.

5. Conclusions

1. In mildly to moderately Cd-contaminated farmlands, the application of lime, compound fertilization, gypsum powder, bamboo charcoal, and optimized fertilization can substantially reduce the effective soil Cd content and rice straw and grain Cd content. They also inhibit the Cd accumulation from the soil to the aboveground rice biomass, thus increasing the rice yield from 4.80% to 14.27%, resulting in the rice grain meeting national standards. The rice grain Cd content was reduced by 23.53% to 36.84%, with optimized fertilization having the most beneficial effects. After optimized fertilization treatment, the cadmium content of rice was the lowest, and the food safety level increased to the highest;
2. The after effect of lime allowed the wheat from the mildly to moderately Cd-contaminated land to meet food safety standards. The compound passivator, gypsum powder, bamboo charcoal, and optimized fertilization all had a fixed action time and limited capacity. The after effect restricted the wheat grain Cd content from meeting national standards. Therefore, the additional application of passivation or optimized fertilization after the second cultivation season is recommended;
3. A comprehensive comparison between the safe utilization and economic effects of lime, compound passivator, gypsum powder, bamboo charcoal, and optimized fertilization showed that optimized fertilization had the optimal outcome. It only requires a one-time application, which improves operation simplicity and saves time and costs. Furthermore, optimized fertilization can improve soil fertility and increase crop yield. Optimized fertilization can facilitate the safe utilization of mildly to moderately Cd-contaminated farmlands, and has excellent economic benefits. Therefore, this research demonstrates it to be suitable as a long-term strategy for reducing rice Cd uptake.

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