Ecotoxicology and Environmental Safety 256 (2023) 114884

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv

Different composites inhibit Cd accumulation in grains under the rice-oilseed rape rotation mode of karst area: A field study

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ARTICLE INFO

Edited by Dr. Muhammad Zia-ur-Rehman

Keywords:
Cadmium
Paddy soil
Composite amendments
Paddy-upland rotation
Total cost

ABSTRACT

Ensuring the safe production of food and oil crops in soils with elevated cadmium (Cd) content in karst regions is crucial. We tested a field experiment to examine the long-term remediation effects of compound microorganisms (CM), strong anion exchange adsorbent (SAX), processed oyster shell (POS), and composite humic acids (CHA) on Cd contamination in paddy fields under a rice-oilseed rape rotation system. In comparison to the control group (CK), the application of amendments significantly increased soil pH, cation exchange capacity (CEC), and soil organic matter (SOM) content while markedly decreasing the content of available Cd (ACd). During the rice cultivation season, Cd was predominantly concentrated in the roots. Relative to the control (CK), the Cd content in each organ was significantly reduced. The Cd content in brown rice decreased by 19.18–85.45%. The Cd content in brown rice following different treatments exhibited the order of CM > POS > CHA > SAX, which was lower than the Chinese Food Safety Standard (GB 2762–2017) (0.20 mg/kg). Intriguingly, during the oilseed rape cultivation season, we discovered that oilseed rape possesses potential phytoremediation capabilities, with Cd mainly accumulating in roots and stems. Notably, CHA treatment alone significantly decreased the Cd content in oilseed rape grains to 0.156 mg/kg. CHA treatment also maintained soil pH and SOM content, consistently reduced soil ACd content, and stabilized Cd content in RSF within the rice-oilseed rape rotation system. Importantly, CHA treatment not only enhances crop production but also has a low total cost (1255.230 US $\$/hm^2$). Our research demonstrated that CHA provides a consistent and stable remediation effect on Cd-contaminated rice fields within the crop rotation system, as evidenced by the analysis of Cd reduction efficiency, crop yield, soil environmental change, and total cost. These findings offer valuable guidance for sustainable soil utilization and safe production of grain and oil crops in the context of high Cd concentrations in karst mountainous regions.

1. Introduction

The contamination of cultivated lands with cadmium (Cd) is a significant ecological issue (Li et al., 2021), particularly in paddy fields (Zhang et al., 2022). The high toxicity and widespread pollution associated with Cd (Ullah et al., 2021) not only affect food security but also pose a threat to human health (Li et al., 2022a; McLaughlin et al., 2021; Yang et al., 2022a). In China, regions with soil contamination by Cd are predominantly found in the central, southern, and southwestern parts of the country (Hu et al., 2020; Zou et al., 2021). Furthermore, the level of Cd pollution in rice (*Oryza sativa* L.)-producing areas in southern China has been increasing each year (Yang et al., 2021). Soil Cd contamination

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https://doi.org/10.1016/j.ecoenv.2023.114884

Available online 11 April 2023

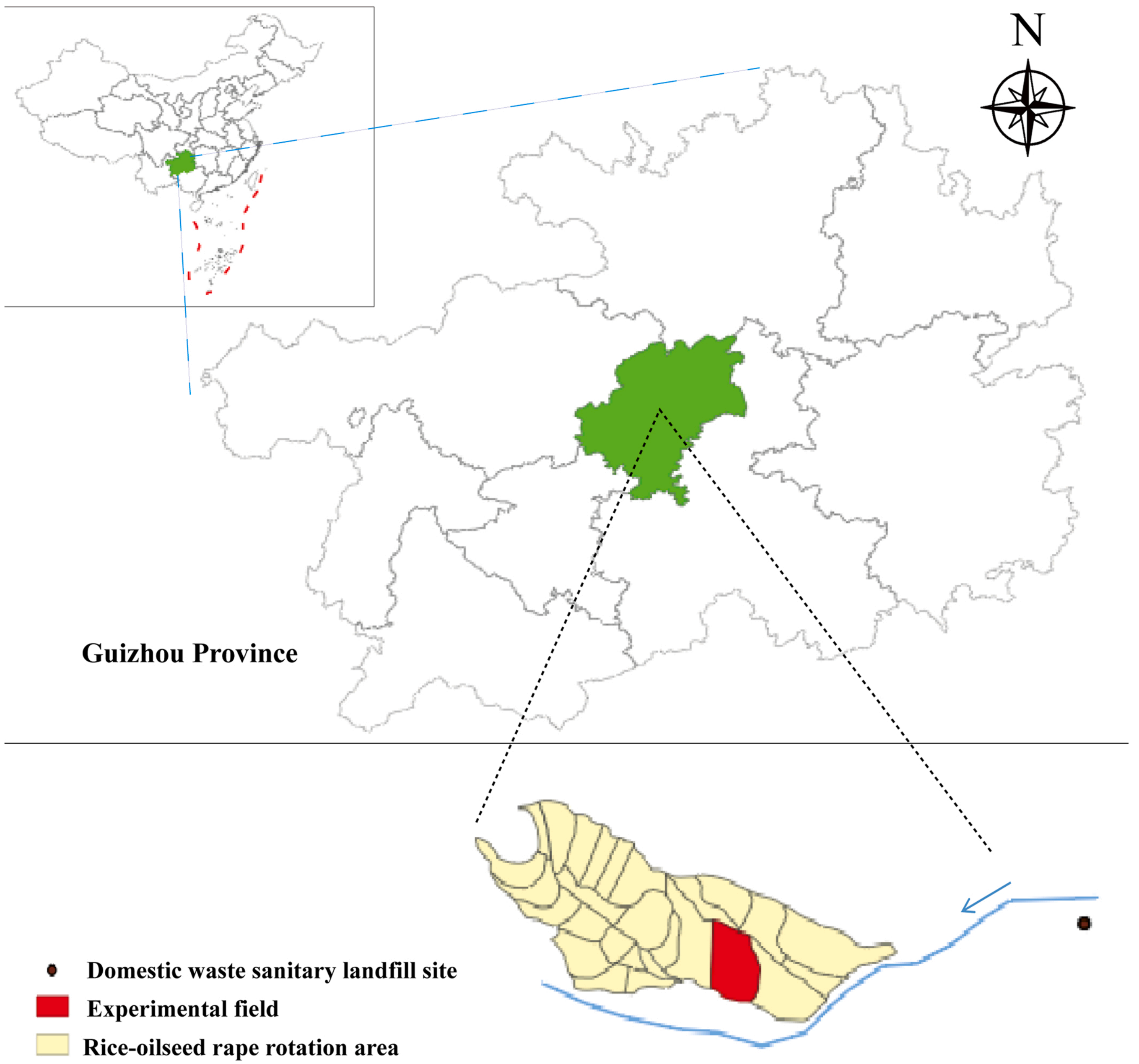

Fig. 1. Schematic diagram of the geographical location of the experimental field.

in typical karst regions is primarily derived from the parent material of the soil (Zhang et al., 2022). As a result, the Cd content (0.659 mg/kg) of cultivated lands in Guizhou exhibits high geological background values (Wen et al., 2020). Hence, addressing soil Cd contamination is essential for guaranteeing the sustainable utilization of agricultural land and the safe production of food.

The use of remediation materials such as clay minerals (Ma et al., 2021), composite humic acids (CHA) (Mosa et al., 2021), and bio-improved modifiers (He et al., 2021) represents some of the most direct and effective approaches for treating Cd-contaminated soil (Hannan et al., 2021). The toxicity and mobility of Cd are contingent on the total content of its specific form and binding state (Hamid et al., 2019; Hamid et al., 2020). Previous research has shown that strong anion exchange adsorbents, with numerous pores and exceptional adsorption capacity, can considerably decrease Cd enrichment, foster rice growth, and preserve soil health (Hamid et al., 2019; Ma et al., 2021). The study demonstrated that after employing nanocomposites, the adsorption efficiency of Cd (II) ions could achieve by over 93% (Awual et al., 2018). Furthermore, the adsorption of metal ions by variable charge soils and minerals intensifies as their pH, clay, and organic matter (OM) contents increase (Naidu et al., 1997). CHA can lead to a decrease in the concentration of Cd in oilseed rape (*Brassica napus* L.). This, in turn, can enhance crop yields by reducing the exchangeable fraction of Cd (Zhou et al., 2018). He et al. (2021) reported that bioimprovement techniques could effectively enhance soil pH levels and decrease the activity of Cd in the rhizosphere of variable charge soils. These improvements are primarily achieved through various mechanisms such as ion exchange, precipitation, and complexation. Additionally, the accumulation of Cd in rice can be reduced as a result of these interventions. Furthermore, the use of composite microbial agents can help to mitigate the mobility and bioavailability of Cd in soil by converting mobile forms of Cd into more stable forms (Kong and Lu, 2022). In a previous study, it was reported that an increase in the bacterial population of variable charge soil could promote the

Table 1
Characteristics and application amount of the amendments used in the present study.

Treatments	Amendments	Main components in passivators	pH	Application rate (kg/hm^2)	Company
CK	Control	-	-	-	-
CM	Complex microorganisms	Brevibacillus lateral spore, Bacillus licheniformis, bioactive calcium, organic matter carrier, total N:0.38%, total P:0.88%, total K:1.99%	8.98	3000	Beijing Century Arms Biotechnology Co., LTD
SAX	Strong anion exchange adsorbent	SiO_2:1.6% ; MgO : 4.49% ; CaO : 6.99%, total N:0.91%, total P:0.10%, total K:0.35%	12.15	6000	Gefeng Technology Materials Co., LTD
POS	Processed oyster shell	CaO ≥ 45%,oyster shell powder, total N:0.53%, total P:0.12%, total K:0.16%	8.29	3000	Fujian Mata Ecological Technology Co., LTD
CHA	Composite humic acids	Naturally ionized minerals; macromolecule humic acid, total N:0.25%, total P:0.12%, total K:0.27%	8.51	3750	Beijing Yihe Yuan Soil Ecological Technology Co., LTD

CK: control; CM: compound microorganisms; SAX: strong anion exchange adsorbent; POS: processed oyster shell; CHA: composite humic acids.

immobilization of Cd^{2+} in the soil matrix. This, in turn, results in a significant reduction in the concentration of Cd in the root and stem tissues of plants (Ali et al., 2022). In a previous study, we demonstrated that the use of ternary compounds in combination could lead to more effective remediation of Cd-contaminated soil in rice fields compared to the use of single materials. This was observed through a pot experiment (Wei et al., 2019). The findings of this study indicate that further research is needed to investigate the long-term remedial effects of the amendments applied in actual field crop production. Moreover, the total cost of such interventions should also be taken into consideration.

The rice-oilseed rape rotation planting system is commonly used worldwide (Huang et al., 2021), and these crops are major sources of grain and oil production in China. Rice is a staple food for more than half of the global population (Li et al., 2021). Furthermore, oilseed rape is a promising hyperaccumulator plant that can be utilized for the restoration of Cd-contaminated soil (Wu et al., 2021). Earlier studies have indicated that crop rotation involving oilseed crops can effectively facilitate the removal of Cd from soil (Yang et al., 2022). Currently, the majority of studies focused on the remediation of Cd-contaminated soil using amendments employ simulation experiments (Li et al., 2022b), pot experiments (Huang et al., 2021), or single-season planting models (He et al., 2021a; Liu et al., 2021). While these studies have demonstrated a certain degree of efficacy in reducing Cd levels, it is still unclear whether these interventions can lead to sustained and stable remediation effects in subsequent crop cycles.

The objective of this study was to examine the ongoing remediation impact of various composite soil amendments on Cd-contaminated soil in the field within the context of a rice-oilseed rape rotation system in karst regions. Initially, we assessed the influence of composite amendments on the soil environment. Subsequently, we investigated the Cd enrichment and absorption mechanisms in crops under the application of composite amendments. Lastly, to encourage the large-scale adoption of these amendments, we analyzed the total cost associated with their use. The findings of this research hold practical significance for advancing sustainable soil use and ensuring the safe production of rice and oilseed rape in areas with high Cd concentrations in karst mountainous regions.

2. Materials and methods

2.1. Test materials

The experimental field (Fig. 1) was located in Kaiyang County (106°44' E, 26°47' N) in Guizhou Province, China. This region is characterized as a typical karst area with a high geological background value of Cd (0.659 mg/kg) (Wang et al., 2020). The region has an annual frost-free period of approximately 315 days, with an average temperature range of 10.6–15.4 °C and an annual precipitation rate of 1150 mm. The complex topography in this region results in strong weathering and dissolution of flowing water. The primary landform type is mountainous terrain which is characteristic of typical karst areas. The soil in the area is classified as paddy soil, and the basic physical and chemical properties of the 0–20 cm soil layer were as follows: pH 6.00, organic matter 43.45 g/kg, exchangeable calcium 1265.81 mg/kg, exchangeable magnesium 249.87 mg/kg, total nitrogen 1.93 mg/kg, available phosphorus 9.15 mg/kg, and available potassium 89.0 mg/kg. The available contents of boron, molybdenum, zinc, manganese, iron, and copper in the soil were 0.48, 0.26, 3.52, 33.76, 171.01, and 2.99 mg/kg, respectively. The total contents of Cd, Hg, As, Pb, Cr, Ni, and Cu in the soil were 1.28, 0.152, 8.54, 71.8, 63.8, 30.2, and 27.4 mg/kg, respectively (Table S1). The average concentration of soil Cd was 1.28 mg/kg, which exceeded the soil pollution risk screening value for agricultural land (GB15618–2018).

Tianyou 1177 is an early-maturing indica three-line hybrid rice variety with a growth period of 157.9 days. The experimental oilseed rape variety used in this study is Xinde Zayou 9, which was bred using 6017A/2006 C (a three-line hybrid) and has a growth period of 227 days. Table 1 presents the basic information about the four amendments utilized in this study. CM is a polymer material synthesized using macrogenomics and high-throughput screening technology. It has a pH of 8.98, and its main components include *Bacillus lateralis, Bacillus licheniformis*, bioactive calcium, and organic carrier. The microbial richness is 200 cfu/mL. SAX comprises of clay, montmorillonite, and a modifier and is a type of nanomaterial characterized by a high specific surface area and a porous structure. It has a pH of 12.15, and its primary components include SiO_2 (1.6%), MgO (4.49%), CaO (6.99%), and a specific surface area of 510 m^2/g. POS utilizes oyster shells as raw material and has a pH of 8.29. The primary component is CaO (≥ 45%), and it features an irregular microporous structure with pore sizes ranging from 2 to 10 μm. CHA is produced from humic acid and minerals by adjusting pressure, alkali consumption, temperature, and time parameters under hydrothermal reaction conditions. It has a pH of 8.51, and its primary components are natural ionized minerals and high molecular humic acid. The humic acid content comprises 23.46% organic carbon, 38.58% free humic acid, 0.21% water-soluble humic acid, 0.23% total phosphorus, and 2.33% total potassium.

2.2. Experiment design

The experiment was carried out from April 2019 to May 2020, with rice planted in the first season from April 2019 to September 2019 and oilseed rape in the second season from October 2019 to May 2020. Five treatments were established, namely: control (CK), composite microorganisms (CM), strong anion exchange adsorbent (SAX), processed oyster shell (POS), and composite humic acids (CHA). Four replicates were implemented for each treatment using a randomized complete block design (Fig. S1). The field plot area was 20 m^2 (4 m × 5 m), and on June 1, 2019, the four amendments were evenly distributed in the tillage layer (0–20 cm) of each plot and only apply them once. Each plot was irrigated and drained independently, and standard field management practices, including fertilization and irrigation, were implemented. Rice formula fertilizer was used as fertilizer, and its total nitrogen,

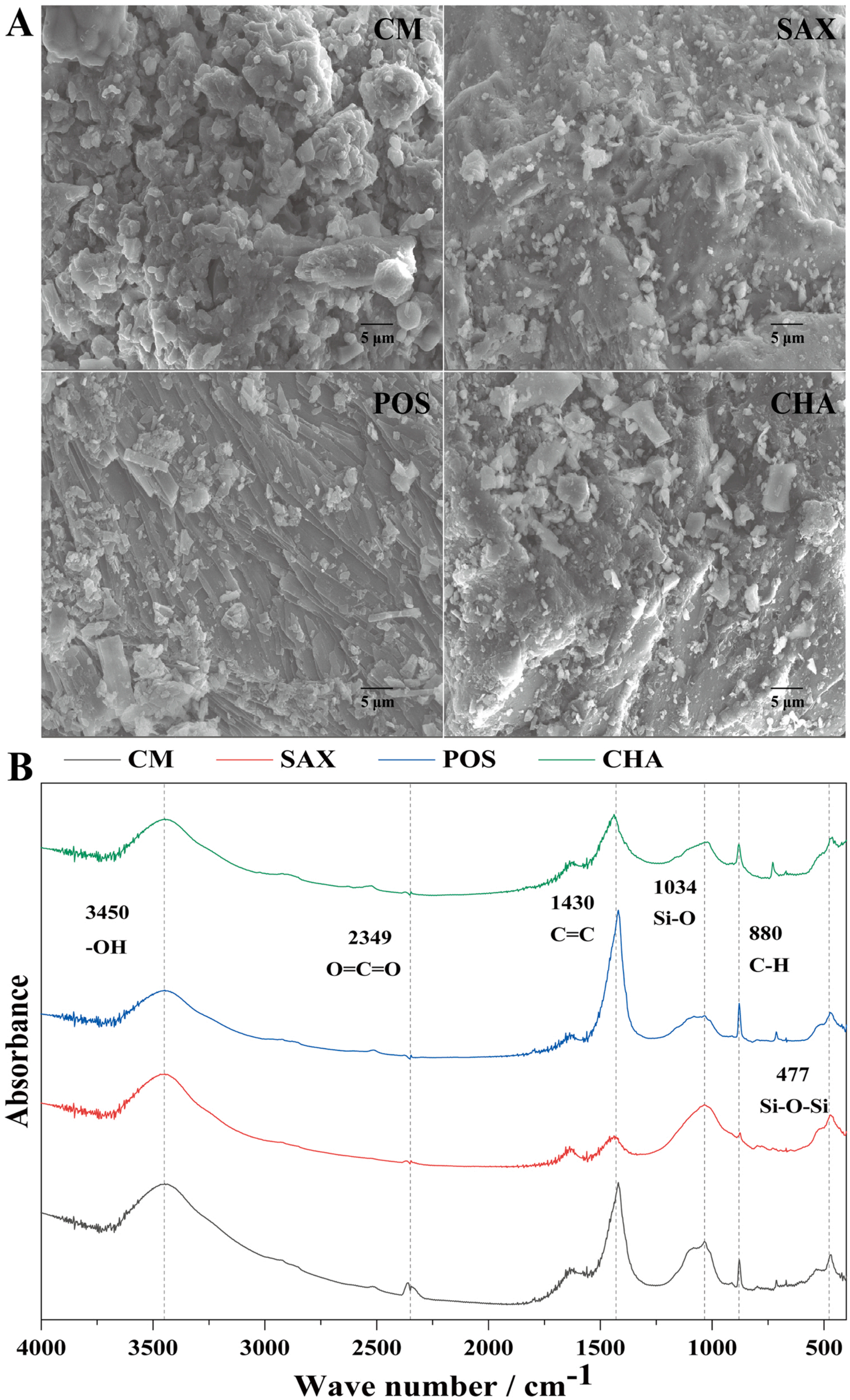

Fig. 2. Scanning electron micrographs of the four amendments (A). The Fourier transform infrared spectra of the four amendments (B). CK: control; CM: compound microorganisms; SAX: strong anion exchange adsorbent; POS: processed oyster shell; CHA: composite humic acids.

phosphorus, and potassium content was 52%, with a proportion of 24:10:18 and an application rate of 600 kg·hm^{-2}. Additionally, to avoid the impact of landfill waste upstream of the irrigation water source, irrigation ditches were built to irrigate with water from Dongfeng Reservoir in Kaiyang County. The rice field was exposed to the sun once during the tillering stage and then drained and dried seven days before harvesting. The planting density of rice was 150,000 plants per hectare. The planting density of oilseed rape was 135,000 plants per hectare. Rice was transplanted on June 8, 2019 (at the three-leaf stage) and harvested on September 28, 2019, while oilseed rape was transplanted

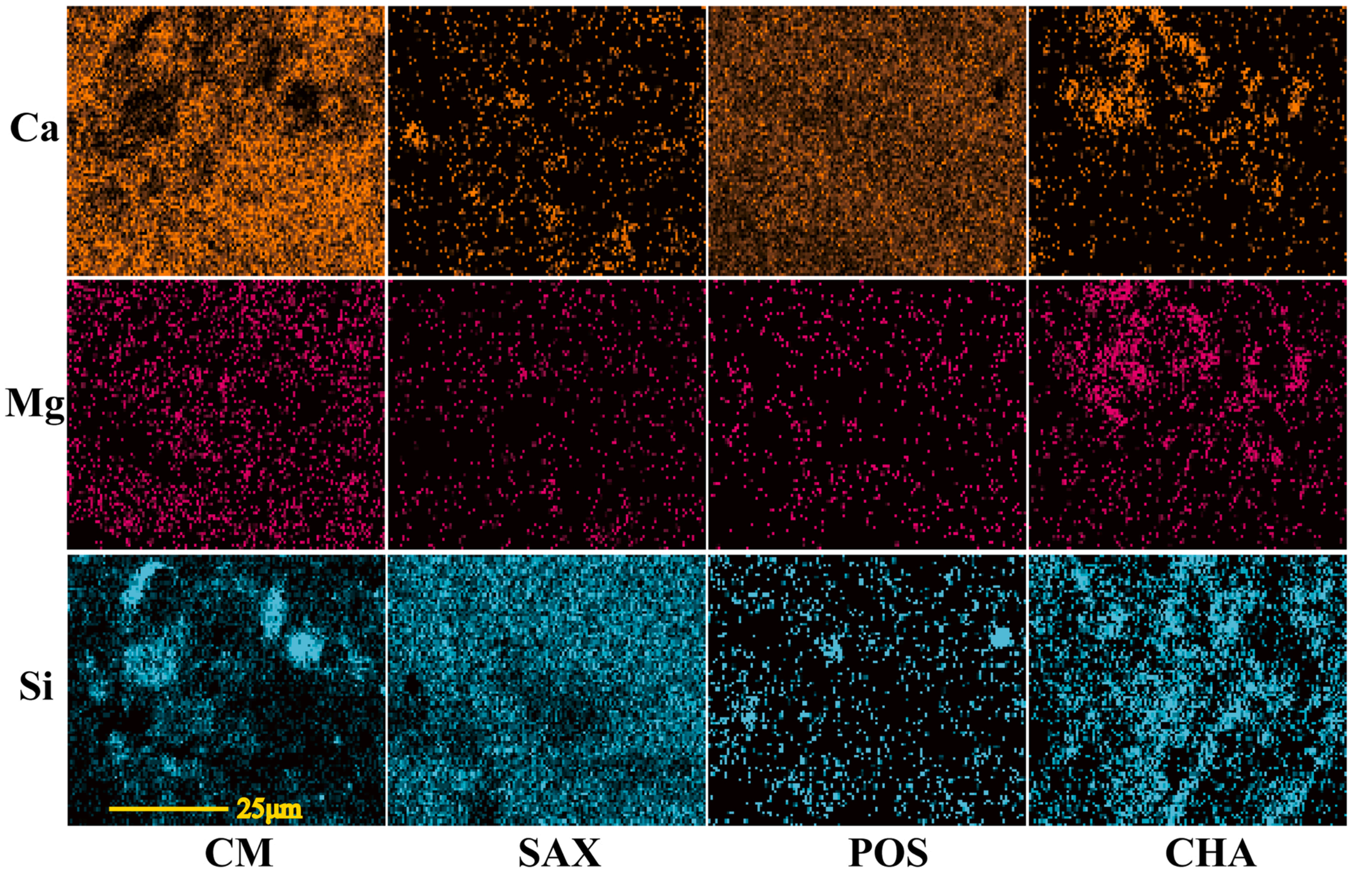

Fig. 3. Ca, Mg, and Si ions on the surfaces of CM, SAX, POS, and CHA. The color difference represents the concentration of ions. CM: compound microorganisms; SAX: strong anion exchange adsorbent; POS: processed oyster shell; CHA: composite humic acids.

on November 6, 2019 (at the three-leaf stage) and harvested on May 27, 2020.

2.3. Soil and plant sampling

The plant-soil samples were collected using the five-point sampling method (Jiang et al., 2022). Soil samples were collected from the cultivated land layer (0–20 cm), while the crops from each plot were harvested separately to measure their yields. Rice plants were washed with both tap and deionized water and were then separated into roots, stems, leaves, and cobs. On the other hand, oilseed rape plants were divided into samples of roots, stems, hulls, and grains. The samples were then dried at 105 °C for 2 h and subsequently at 65 °C until a constant weight was achieved. Later, the samples were ground using a grinding miller (IKA A11, Germany). The soil was air-dried, ground, and successively passed through 10- and 100-mesh nylon screens to prepare soil samples for testing (Li et al., 2020).

2.4. Sample analysis

Morphological observation and elemental analysis of each amendment were performed using scanning electron microscopy (SEM; TESCAN MIRA LMS, UK). Fourier transform infrared spectroscopy (FTIR; Nicolet 6700 FTIR spectrometer, UK) was utilized to analyze the surface functional groups of each amendment (Table S1) (Zhao et al., 2021). Soil pH was determined using the potentiometric method with a soil-to-water ratio of 2.5:1. Total N content in soil samples was analyzed using the Kjeldahl method. The acid fusion-Mo-Sb colorimetric method and flame photometry were utilized to measure total P and K, respectively. Soil organic matter (SOM) content was determined using the potassium thermochromate oxidation-volumetric method at high temperatures. The cation exchange capacity (CEC) of the soil was measured by shaking the soil with 1 mol L^{-1} NH_4Ac (Kermel, Tianjin, China) acetate (Bao, 2000). The method used to determine soil available silicon (ASi) was the molybdenum blue method (Korndörfer et al., 2001). The method of EDTA titration was employed to estimate the soil's exchangeable calcium (EX-Ca) and magnesium (EX-Mg) (Heald, 1965). Soil total Cd (TCd) content was determined by digestion through the high-pressure closed digestion method. Soil available Cd (ACd) content was estimated by extraction with the DTPA method. The content of Cd forms in soil was measured by the improved BCR extraction method (Cui et al., 2016; Zong et al., 2021), which were exchangeable fraction (EXF), a reducible fraction (REF), oxidizable fraction (OXF), and residual fraction (RSF) (Table S2). The Cd content in plant samples was digested with a mixture of HNO_3-H_2O_2 and then determined by inductively coupled plasma mass spectrometry ICP-MS (Thermo Fisher Scientific, USA). The detection limit of the analytical methodology Cd quantification by ICP-MS is 0.001 mg/kg, and the detection line is 0.004 mg/kg. The sensitivity is as per the technical requirements (RSD: 1.05–2.37%), where the linearity is $F(x) = 15{,}032.1414 *x + 328.6873$ ($R^2 = 1.0000$).

The biological concentration factor (BCF) and transport factor (TF) are measures used to evaluate the accumulation and transportation ability of Cd heavy metal in different parts of crops. The calculation of BCF and TF is as follows:

$$BCF = Ci/Cs.TF = C\ latter/C\ former$$

where C_i is the measured content of heavy metal Cd in various parts of crops (mg/kg), C_s is the total Cd content of paddy soil (mg/kg), C_{latter} refers to the heavy metal Cd content (mg/kg) in the latter part, and C_{former} refers to the heavy metal Cd content (mg/kg) in the former part. (Wang et al., 2021).

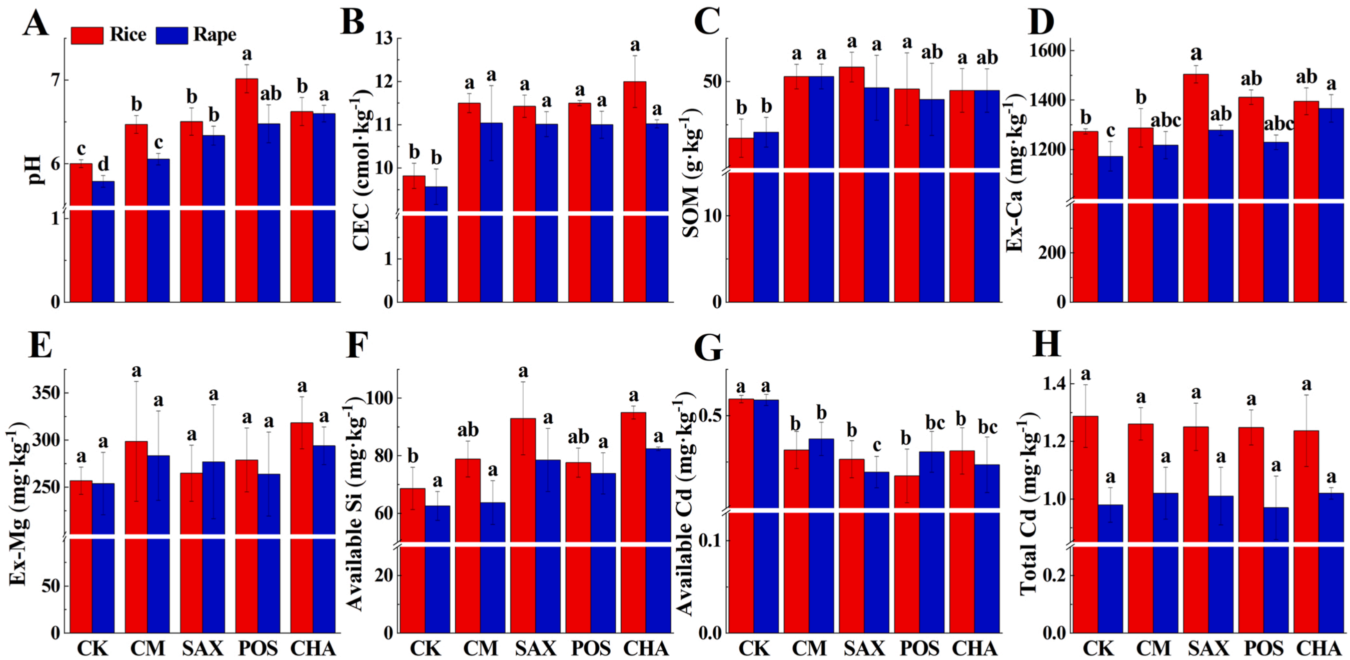

Fig. 4. Effects of the four amendments on soil in the rice-oilseed rape rotation mode: (A) soil pH; (B) CEC:cation exchange capacity; (C) SOM:soil organic matter; (D) EX-Ca:exchangeable calcium; (E) EX-Mg:exchangeable magnesium; (F) ASi:available silicon; (G) ACd:available cadmium; and (H) TCd:total cadmium. The different lowercase letters indicate significant differences among the treatments for crops in the same season ($p < 0.05$). Values in the figure are expressed as mean ± standard error (n = 4). CK: control; CM: compound microorganisms; SAX: strong anion exchange adsorbent; POS: processed oyster shell; CHA: composite humic acids.

2.5. Quality assurance

To maintain the analytical accuracy, standard substances were employed, including soil (GBW07405 (GSS-5), IGGE, Jiangsu, China) and plant (GBW10010 (GSB-1), IGGE, Beijing, China), and the recovery rate of Cd ranged from 91.67% to 100.09%. Additionally, a reference material (GBW07443 (GSF-3), IGGE, Hubei, China) was used to monitor the quality of soil morphological components, and the recovery rate of Cd was between 99.50% and 102.30%.

2.6. Statistical analysis

Data analysis and processing were performed using Microsoft Excel 2019 and SPSS software (Ver. 22.0, IBM, USA). Mapping was carried out using ArcGIS 10.6, Origin (Origin Pro v 9.0, Origin Lab, Northampton, USA 2021), and TBtools (v1.098, South China Agricultural University, Guangdong, China) software (Chen et al., 2020). Multiple comparisons were performed using one-way analysis of variance (ANOVA) and least significant difference (LSD) test, with statistical significance set at $p < 0.05$ and $p < 0.01$.

3. Results

3.1. SEM and FTIR analysis of the four composite amendments

Fig. 2A depicts the microscopic morphological characteristics of the four composite amendments. CM had a porous texture and a loose bulk structure. In contrast, SAX had a dense bulk structure. POS and CHA had a lamellar structure, and a blocky structure, respectively, and both had an irregularly stacked microporous structure. The FTIR spectra of the four amendments, ranging from 400 to 4000 cm^{-1}, exhibited a wide range of surface functional groups containing carbon (C), oxygen (O), silicon (Si), and hydrogen (H) (Fig. 2B). The FTIR spectra also revealed the presence of organic functional groups such as C-H, C =C, and -OH in all four composite amendments. Furthermore, electron microscopic analysis (Fig. 3) revealed that the four composite amendments contained calcium, magnesium, and silicon ions, with CM and POS containing a high concentration of calcium ions, and SAX and CHA containing a large amount of silicon ions.

3.2. Effect of the four treatments on the soil environment in rice-oilseed rape rotation system

In the rice season, all four amendments significantly increased soil pH compared to the CK by 0.47–1.02 units (Fig. 4A), with the POS treatment showing the highest pH value of 7.02. Additionally, the CEC (Fig. 4B) and SOM (Fig. 4C) were significantly ($p < 0.05$) increased by the four amendments compared to CK. The EX-Ca content (Fig. 4D) was also increased by the four amendments, with the SAX treatment having the highest EX-Ca content at 1504.98 $mg \cdot kg^{-1}$. The EX-Mg content (Fig. 4E) was significantly increased by the four amendments compared to CK. Moreover, the SAX and CHA treatments significantly ($p < 0.05$) increased ASi content (Fig. 4F), with the CHA treatment having the highest ASi content of 95.06 $mg \cdot kg^{-1}$. The four amendments significantly reduced the ACd content (Fig. 4G) compared to CK, with the POS treatment showing the lowest ACd content. The total Cd (TCd) content (Fig. 4H) was consistent across all treatments at 1.26 $mg \cdot kg^{-1}$.

In the oilseed rape season, compared to CK, the four soil amendments significantly increased the soil pH ($p < 0.05$), and the increase was 0.27–0.81 units; the soil pH value of CHA treatment was the highest at 6.60 (Fig. 4A). Furthermore, compared to CK, the different treatments significantly reduced soil ACd content ($p < 0.05$), with a reduction of 8.21–15.07%; SAX treatment showed the lowest value with 0.439 $mg \cdot kg^{-1}$ (Fig. 4G). The average soil TCd content was 0.99 $mg \cdot kg^{-1}$ under different treatments (Fig. 4H).

Fig. 4 depicts the changes in soil indices before and after the application of the four amendments in the rice-oilseed rape rotation system. The results showed that the CHA treatment was relatively effective in increasing soil pH within a short period, whereas the SOM content varied slightly following the CM and CHA treatments (Fig. 4B) but decreased after SAX and POS treatments, decreasing by 4.63% and 2.44%, respectively. This finding indicates that CM and CHA treatments

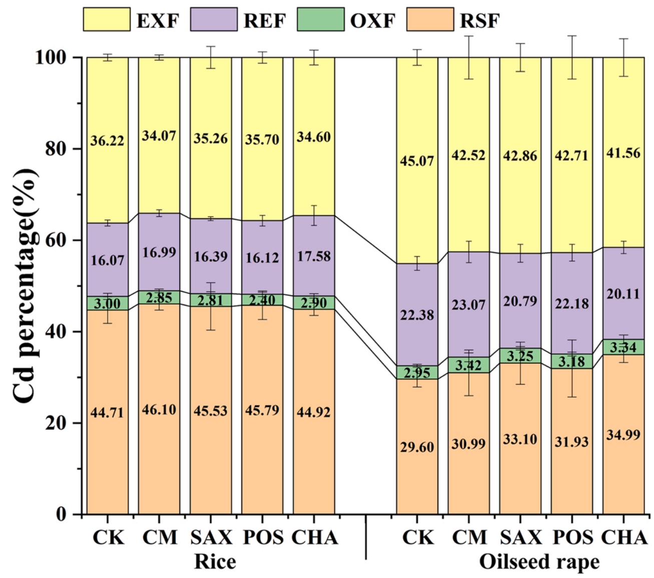

Fig. 5. Effects of four amendments on different forms of soil Cd in rice-oilseed rape rotation system. EXF: exchangeable fraction; REF: reducible fraction; OXF: oxidizable fraction; RSF: residual fraction. Values in the figure are mean ± standard error (n = 4). CK: control; CM: compound microorganisms; SAX: strong anion exchange adsorbent; POS: processed oyster shell; CHA: composite humic acids.

could maintain the relative content of SOM. As shown in Fig. 4C, SAX and CHA treatments reduced soil ACd by 3.11% and 3.37%, respectively. Thus, CHA treatment could maintain soil pH and SOM content and continuously reduce soil Cd activity in the rice-oilseed rape rotation system.

3.3. Changes in Cd chemical fraction

To investigate the cause of the decrease in soil TCd content in the oilseed rape season compared to that in the rice season, the changes in soil Cd morphological content in the mature stage of rice and oilseed rape were measured (Fig. 5). During the rice season, RSF was the primary form of Cd occurrence (Fig. 5A), with the order of Cd occurrence in the soil being RSF > EXF > REF > OXF. Compared to CK, the four treatments reduced the EXF content of soil Cd in the rice season to varying degrees while the REF content increased. However, all treatments reduced the OXF content in the soil, with the proportion in the total amount of Cd being the lowest. Furthermore, compared to CK, the soil RSF content increased by 2.48–5.63%. This suggests that the four treatments reduced the EXF content of soil Cd, thus, increasing the RSF content.

In contrast, during the oilseed rape season, EXF was the main Cd form occurring in the soil (Fig. 5B). Compared to CK, the EXF content in soil treated with the four amendments decreased, with a percentage decrease ranging from 1.67% to 6.13%. The CM treatment increased the REF content of soil by 7.37%, while in the other treatments, the REF content was reduced by 1.27–4.11%. The OXF content of soil increased by 5.21–19.99%. Moreover, compared to CK, the soil treated with the four amendments showed an increase in RSF content by 7.02–26.21%, and CHA treatment significantly increased RSF content ($p < 0.05$). These results suggest that CHA treatment increases RSF content and subsequently continuously decreases soil ACd content.

3.4. Cd content in rice and oilseed rape organs

The Cd content in various organs of rice and oilseed rape plants was reduced by the four amendments compared to the control treatment (Fig. 6). During the rice season, the Cd content of rice roots, stems, leaves, cobs, rice husks, and brown rice was significantly reduced ($p < 0.05$) by 33.90–70.75%, 35.22–94.04%, 60.21–73.58%,

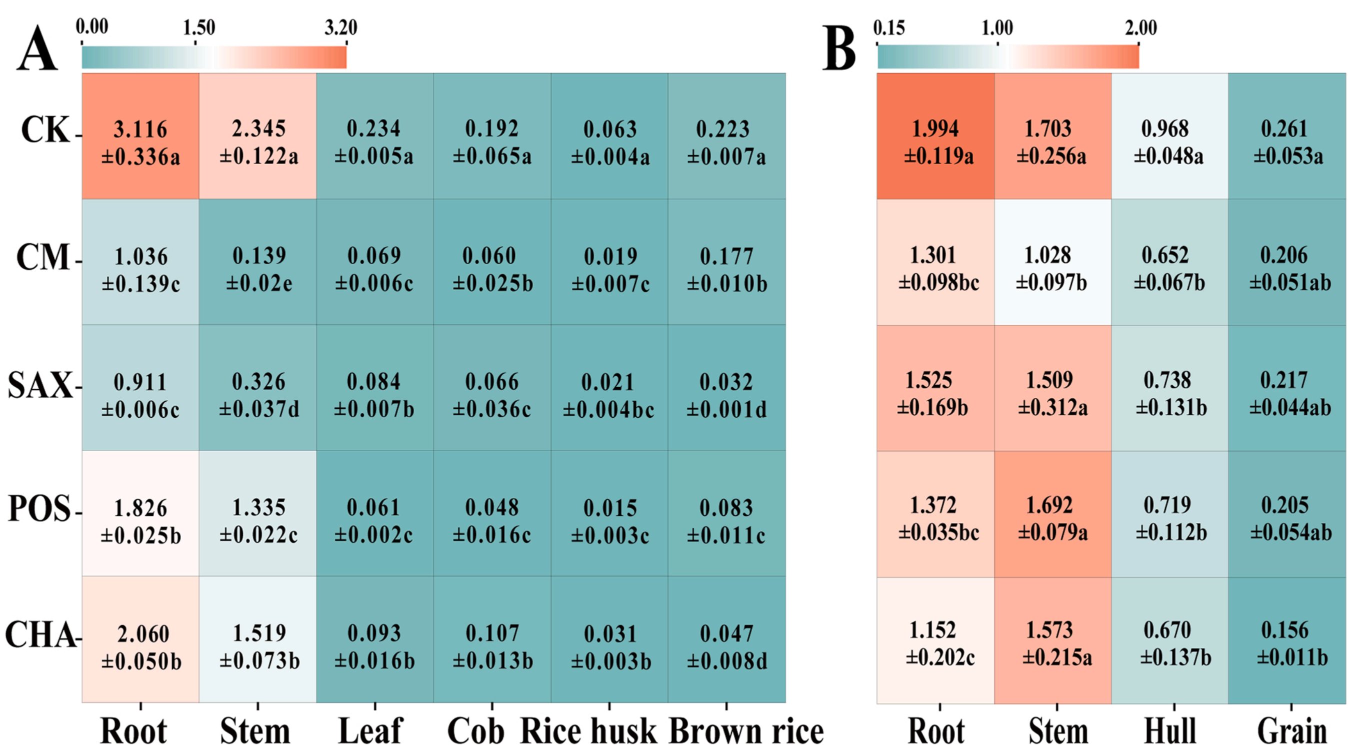

Fig. 6. Cd distribution in the various organs of crops under different amendments. The color difference indicates the level of Cd content ($mg \cdot kg^{-1}$). Values in the figure are expressed as mean ± standard error (n = 4). The different lowercase letters in the same column represent a significant difference ($p < 0.05$) between the treatments. CK: control; CM: compound microorganisms; SAX: strong anion exchange adsorbent; POS: processed oyster shell; CHA: composite humic acids. (A) represents rice season; (B) represents oilseed rape season.

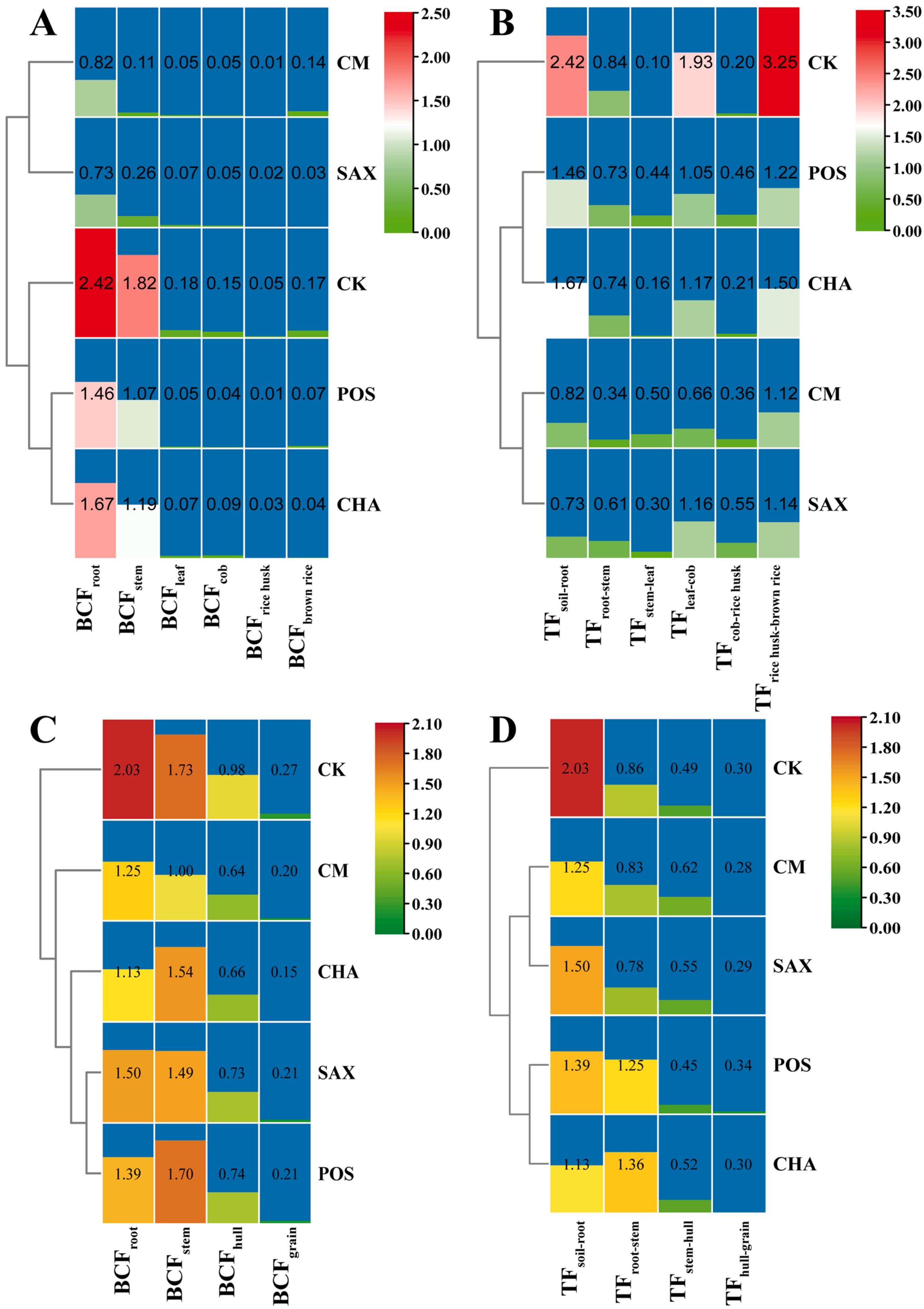

Fig. 7. Effects of the four amendments on Cd enrichment and absorption in the organs of rice and oilseed rape. Values in the figure are expressed as mean ± standard error (n = 4). CK: control; CM: compound microorganisms; SAX: strong anion exchange adsorbent; POS: processed oyster shell; CHA: composite humic acids. (A) shows the enrichment coefficients of rice organs. (B) shows the transport coefficients of rice organs. (C) shows the enrichment coefficients of oilseed rape organs. (D) shows the transport coefficients of oilseed rape organs.

43.99–74.93%, 50.39–76.85%, and 19.18–85.45%, respectively, when treated with different amendments compared to the control (Fig. 6A). The Cd content in brown rice was lower than the maximum allowable limit of 0.2 mg·kg^{-1} (GB2762–2017), and the order of Cd content after different treatments was CM > POS > CHA > SAX.

In the oilseed rape season, the Cd contents in roots, stems, shells, and grains were significantly reduced by 23.48–42.22%, 0.65–39.65%, 23.76–30.75%, and 16.91–40.30%, respectively, under different treatments (Fig. 6B). The reductions in Cd content in roots and shells were significant compared to CK. In grains, the highest decrease in Cd content was observed in the CHA treatment, followed by POS, CM, and SAX treatments. The Cd content in grains treated with CHA was not significantly different from that in the CK treatment ($p < 0.05$), and it was lower than the limit of 0.5 mg·kg^{-1}. Therefore, all four amendments were effective in reducing Cd content in rice and oilseed rape, and CHA treatment was particularly effective in reducing the Cd content of brown rice and oilseed rape grains ($p < 0.05$).

3.5. Enrichment and transport of Cd in crops

The present study revealed that the four treatments significantly decreased the Cd enrichment factor of all parts of rice (Fig. 7A). Specifically, compared to CK, the different treatments markedly reduced the Cd enrichment factor of rice roots and stems ($p < 0.05$), and the lowest enrichment factor of rice roots was observed in the SAX treatment. Additionally, the Cd enrichment factor of rice roots, stems, leaves, cobs, rice husks, and brown rice was reduced by 31.17–69.88%, 34.46–93.91%, 58.57–72.76%, 41.68–74.15%, 48.34–76.13%, and 18.37–84.90%, respectively. Furthermore, the four treatments significantly decreased the Cd enrichment factor of leaves and brown rice ($p < 0.05$) as compared to CK, and the lowest Cd enrichment factor was observed in the SAX treatment. The Cd enrichment factor of cob and rice husk was also significantly reduced after treatment with different amendments as compared to that for CK, and the lowest Cd enrichment factor was observed in the POS treatment. Notably, the order of the enrichment factor of rice organs after CK and CHA treatments was as follows: $BCF_{root} > BCF_{stem} > BCF_{leaf} > BCF_{brown\ rice} > BCF_{rice\ husk}$. The Cd enrichment factor of rice organs after CM, SAX, and POS treatments was as follows: $BCF_{root} > BCF_{stem} > BCF_{brown\ rice} > BCF_{leaf} > BCF_{rice\ husk}$. This indicates that Cd in rice is mainly enriched in roots.

The four amendments showed some differences in the Cd transport capacity of the different parts of rice (Fig. 7B). The TF of POS and CHA treatments were consistent with that of CK treatment, and the order of size was as follows: $TF_{rice\ husk\text{-}brown\ rice} > TF_{leaf\text{-}cob} > TF_{root\text{-}stem} > TF_{cob\text{-}rice\ husk} > TF_{stem\text{-}leaf}$. Compared to CK, the four amendments significantly reduced the TF of leaf to cob ($TF_{leaf\ -\ cob}$) and rice husk to brown rice ($TF_{rice\ husk\ -\ brown\ rice}$) by 39.66–65.91% and 53.86–65.49%, respectively. The $TF_{rice\ husk\ -\ brown\ rice}$ values were greater than 1 in all treatments, which indicates a strong transfer of Cd from rice husk to brown rice.

In the oilseed rape season, the four amendments were found to decrease the Cd enrichment factor in all parts of the plant as compared to CK, with reductions ranging from 25.82% to 44.37% in roots, stems, shells, and grains (Fig. 7C). Obviously, the Cd enrichment factor in the stems and seeds of oilseed rape was significantly reduced. The Cd enrichment factor in the roots and husks of oilseed rape was also significantly decreased ($p < 0.05$), with the lowest enrichment factor observed in the seeds of oilseed rape treated with CHA. The order of the BCF values following CK, CM, and SAX treatments was $BCF_{root} > BCF_{stem} > BCF_{shell} > BCF_{grain}$, whereas, in POS and CHA treatments, the order was $BCF_{stem} > BCF_{root} > BCF_{shell} > BCF_{grain}$. This suggests that Cd is mainly enriched in the roots and stems of oilseed rape.

The Cd translocation factor from soil to roots was in the order of CK > SAX > POS > CM > CHA by the four treatments(Fig. 7D), which significantly decreased the transport capacity of Cd in the soil to the roots of oilseed rape ($p < 0.05$). The order of the Cd translocation factor from root to stem was CHA > POS > CM > SAX, and the CM and SAX treatments reduced the transport factor by 2.76% and 9.73%, respectively. The order of Cd translocation factor from the stem to the shell was CM > SAX > CHA > POS, and the CM and CK treatments reached a significant level. The order of the Cd translocation factor from shell to grain was POS > CHA > SAX > CM. Compared with CK, the POS treatment increased by 10.11%, while the other treatments reduced the factor by 1.60–6.43%. Overall, the $TF_{soil\text{-}root}$ values under the four amendments were all greater than 1, indicating that Cd transport from soil to canola roots is the strongest. The amendments primarily prevent the absorption of soil Cd by roots and the transport of shell to grains, thus, reducing Cd accumulation in grains.

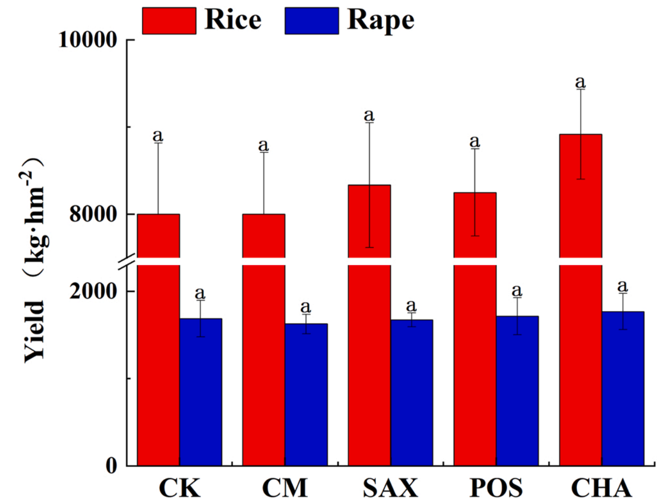

Fig. 8. Effects of the four amendments on the yields of crops. Values in the figure are expressed as mean ± standard error (n = 4). CK: control; CM: compound microorganisms; SAX: strong anion exchange adsorbent; POS: processed oyster shell; CHA: composite humic acids.

3.6. Crop yields

The effects of the four amendments on rice yield were analyzed and compared to CK, and the results are shown in Fig. 8. The rice yield of CK was 8000.04 kg·hm^{-2}, and the yield increased by 3.13–11.46% with the application of CM, SAX, POS, and CHA, with CHA treatment showing the highest yield of 8916.71 kg·hm^{-2}. In contrast, the oilseed rape yield was affected differently by the four amendments, with CM and SAX treatments reducing the yield by 3.59% and 0.84%, respectively, and POS and CHA treatments increasing the yield by 1.58% and 4.91%, respectively. Among these treatments, CHA showed the highest yield of 1770.41 kg·hm^{-2}. These findings indicate that CHA treatment can increase crop yield, while the effects of the other amendments on oilseed rape yield need further investigation.

3.7. Multi-factor correlation analysis

In the rice season (Fig. 9A), our results indicated that the Cd content of brown rice had a significant negative correlation with the levels of ASi and EX-Ca ($p < 0.05$). Moreover, we observed a significant negative correlation between soil pH and ACd ($p < 0.05$). However, ACd was found to be positively correlated with Cd content in rice husks ($p < 0.01$). Additionally, the results showed that the content of reducible Cd fraction (REF) was positively correlated with EX-Mg ($p < 0.05$).

In the oilseed rape season (Fig. 9B), we found that CEC was significantly negatively correlated with ACd ($p < 0.05$). The Cd content in grains was also negatively correlated with EX-Ca and RSF content ($p < 0.05$). However, EX-Ca was positively correlated with RSF content ($p < 0.01$), while EX-Mg was positively correlated with OXF content

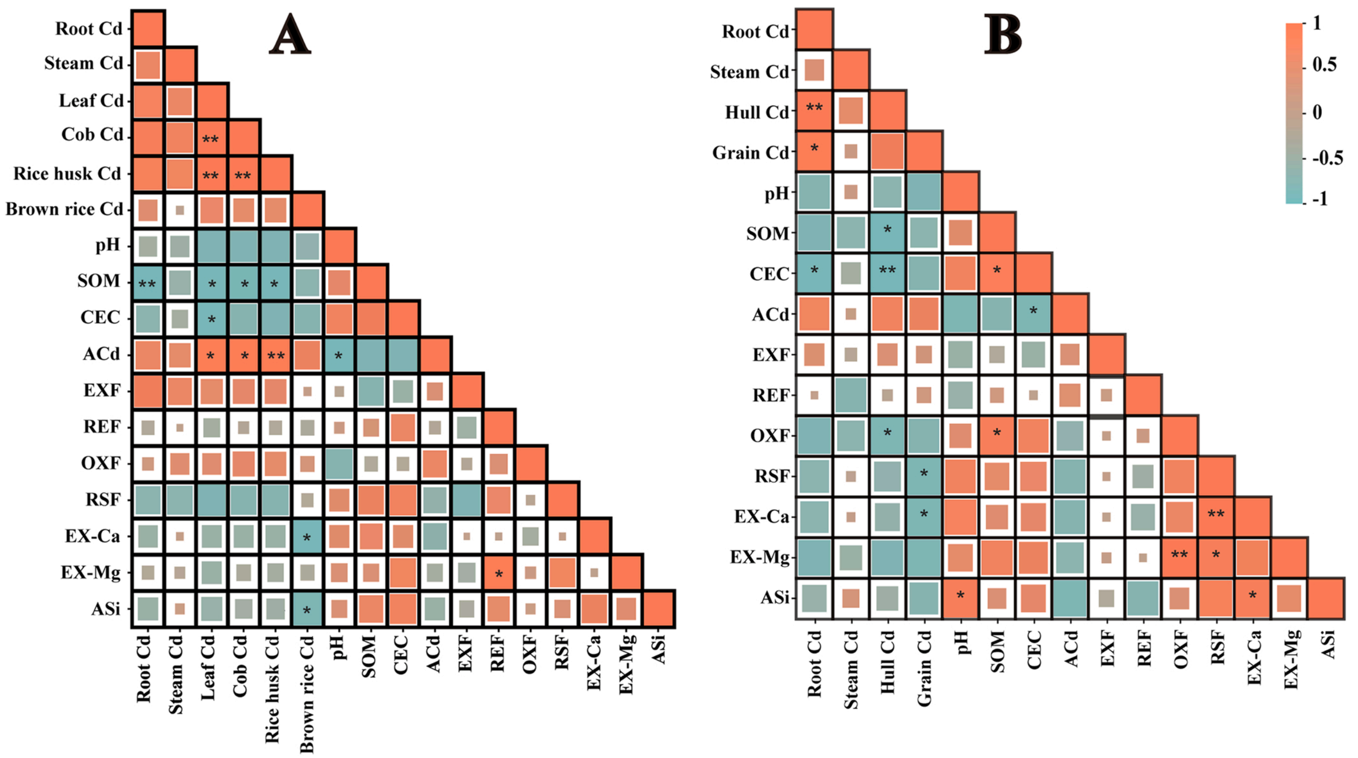

Fig. 9. Correlation analysis of soil and plant indices. The asterisks show a significant correlation. The color difference and the number represent the correlation coefficient magnitude. * : $p < 0.05$; **: $p < 0.01$. (A) represents the rice season; (B) represents the oilseed rape season.

($p < 0.01$). Overall, although EX-Ca was negatively correlated with brown rice and grains ($p < 0.05$), EX-Mg content was positively correlated with RSF content ($p < 0.05$) in the rice-oilseed rape rotation system.

4. Discussion

4.1. Effects of different treatments on soil environment in rice-oilseed rape rotation

The purpose of adding adsorption materials was to increase the amount of Cd adsorbed by the soil. The Cd(II) adsorbed on soils was mainly bound to metal (hydro)oxides or existed as a $CdCO_3$ precipitate (Fan et al., 2022; Ji et al., 2022). Previous studies have indicated that the inclusion of CaO and MgO components facilitates the hydrolysis of Cd^{2+} in the soil to $Cd(OH)^+$ ($Cd^{2+}+H_2O \rightarrow Cd(OH)^+ + H^+$). $Cd(OH)^+$ ions have a higher affinity to soil adsorption sites compared to Cd^{2+}, which decreases the mobility of Cd in soil (He et al., 2021, 2021a). The data presented in Fig. 4 indicate that the four treatments resulted in a significant increase in the content of EX-Ca, EX-Mg, and ASi in the soil as compared to the CK. Interestingly, the correlation analysis presented in Fig. 9 revealed that EX-Ca was found to be negatively correlated with the Cd content in brown rice and grains. This finding suggests that the increased content of EX-Ca could potentially reduce the uptake of Cd by crops, which is consistent with our previous study conducted on potato (*Solanum tuberosum*) (Gong et al., 2022). Recently, it was reported that high concentrations of Ca^{2+} and Mg^{2+} can significantly reduce the accumulation of Cd in various parts of rice (Li et al., 2022c; Okazaki et al., 2008).

Increasing the content of antagonist elements and reducing the translocation and enrichment of Cd in crops can be achieved through the addition of mineral nutrients, which helps prevent Cd toxicity in crops (Qin et al., 2020). In this experiment, both SAX and CHA treatments were observed to reduce Cd enrichment in various parts of rice and rapeseed (Fig. 1), which is likely attributed to the presence of Si in the materials used (Fig. 3). Si is known to play a role in reducing the transport and enrichment of Cd in rice through physiological mechanisms (Liu et al., 2013), such as participating in the physiological metabolic activity of rice (Guo et al., 2022a; Huang et al., 2021b). Cai et al. (2020) demonstrated that applying Si at the jointing stage can effectively decrease the Cd content in brown rice. In addition, Si can enhance crop growth and development by regulating the transport and distribution of Cd as well as the antioxidant enzyme systems in plant tissues (Wu et al., 2018). Additionally, silicon ions can also decrease the exchangeable cadmium in soil and increase the proportion of cadmium bound to carbonates or present in a residual form (Liu et al., 2023). Exogenous application of silicon can facilitate the replenishment of plant-available silicon in soil (Cai et al., 2022). Cd can form a Si-Cd complex with -O-Si-O-, which cannot be enriched by plants. This complex reduces the available cadmium content in soil and hence, reduces the toxicity of Cd to plants (Guo et al., 2022b).

4.2. Inhibition of Cd ions by different amendments in crops

Our findings indicate that the application of CM, SAX, POS, and CHA treatments resulted in a significant reduction in the Cd content of rice organs, with brown rice containing less than 0.2 $mg \cdot kg^{-1}$ Cd (Fig. 6A). Additionally, the Cd content in roots and hulls was significantly reduced during the oilseed rape season (Fig. 6B). Our study is based on the theoretical basis of soil pH regulation for the remediation of Cd-contaminated farmlands (Hussain et al., 2021; Zhao and Wang, 2020). Previous studies have demonstrated that increasing soil pH can lead to a decrease in the concentration of ACd content (Xu et al., 2021a, 2021). All the amendments used in this study were characterized by an alkaline nature, with pH values ranging from 8.29 to 12.15 (Table 1). As compared to CK, the four amendments significantly increased the soil pH (Fig. 4A) and decreased the ACd content (Fig. 4G). Our correlation analysis indicated that soil pH was negatively correlated with ACd content (Fig. 9A). Furthermore, the application of the alkaline SAX was found to increase the soil pH and significantly reduce the ACd content,

Table 2
Comprehensive analysis of the four amendments.

Ranking	Treatment	The total rate of Cd (%)	The total yield rate (%)	Application amount (kg/hm2)	Total cost (US $/hm2)	Product cost (US $/hm2)	Application cost (US $/hm2)
1	CHA	118.22	16.37	3750.0	1255.230	1046.025	209.205
2	POS	83.35	4.72	3000.0	1004.184	836.820	167.364
3	SAX	102.07	3.33	6000.0	1589.958	1255.230	334.728
4	CM	40.15	-3.59	3000.0	1422.594	1255.230	167.364

which effectively inhibited Cd absorption by various organs of rice and oilseed rape (Ma et al., 2021). Interestingly, we found a decrease in total soil Cd content in the root zone in the course of the rape season (Fig. 4H). We speculated that oilseed rape plants had phytoremediation potential. The reason may be that the plant biomass of oilseed rape is larger and it has higher enrichment effect (Tang et al., 2020). Wu et al. (2021) showed that oilseed rape is a potential superenrichment plant to restore Cd-contaminated soil. In order to further verify the reason, we measured the changes of soil Cd morphological content in the mature stage of rice and rape (Fig. 5). The application of the composite amendments reduced the EXF content of soil Cd and increased the RSF content, which may be caused by their special structure (Fig. 2A). The four conditioner materials contain obvious organic functional groups such as -OH, C═C and Si-O (Fig. 2B), and these organic functional groups may enhance the adsorption and chelation of Cd. Meanwhile, Mosa et al. (2021) have revealed humic acid is the microstructure of rigid particles in a compressed shape, containing a variety of oxygen-containing functional groups. After the application of CHA, Cd can bind to more sites such as amines, hydroxyl groups, carboxyl groups, and carbonyl groups through complexation, and the aromatic ring of humic acid promotes cation-π interactions and can effectively fix Cd (Wang et al., 2020). In order to further advance the field, it is recommended that future studies incorporate molecular marker technology (Yan et al., 2019; Yu et al., 2022) in conjunction with agricultural practices like water management (Zhong et al., 2021) to produce and cultivate low Cd-accumulating crop varieties.

4.3. Comprehensive analysis

If the composite amendments can be widely adopted, their effectiveness in reducing Cd accumulation should be considered as a key factor, along with their economic cost (Liu et al., 2021; Meng et al., 2019; Tang et al., 2016; Wang et al., 2022). Previous studies have typically only focused on the product cost of soil amendments rather than considering the cost of application. In our study, we calculated the total costs of CM, SAX, POS, and CHA treatments to be US $ 1422.594, 1589.958, 1004.184, and 1255.230 per square kilometer, respectively, with an average cost of US $ 1317.992 per square kilometer (Table 2). By considering the Cd reduction rate, yield increase rate, and total cost (Table 2), our results suggest that CHA may be a promising option for remediating Cd-contaminated acid rice fields. Our comprehensive evaluation approach offers a new perspective for promoting and applying soil amendments, and future research should focus on improving the evaluation system for soil amendments.

The product cost is based on the market quotation of the product (the development cost is considered if the product is in the experimental stage and has not been introduced into the market). The application cost is calculated based on the cost borne by the farmers for transporting the amendments to the field and the labor cost of the application. The application cost of the amendment was calculated as US$0.558 per hectare per 1000 kg dosage. The total reduction rate of Cd (%) was calculated as follows: Cd reduction rate by brown rice (%) + Cd reduction rate by oilseed rape (%). The total yield rate (%) was determined as follows: Rice yield increase rate (%) + Oilseed rape yield increase rate (%). The total cost was calculated as follows: Product cost + Application cost.

5. Conclusions

After applying the four amendments, there was a significant increase in soil pH, CEC, and SOM content, while the ACd content was reduced. In the rice season, Cd was mainly enriched and transported in the roots. All treatments significantly reduced the Cd content in each organ, with the lowest Cd content in brown rice, ranging from 19.18% to 85.45% lower than CK. The order of Cd reduction efficiency in brown rice was CM > POS > CHA > SAX, and all treatments met the Chinese Food Safety Standard for Cd content in grains (0.20 mg/kg). In the oilseed rape season, Cd was mainly enriched in the roots and stems. CHA alone significantly reduced Cd content in oilseed rape grains to 0.156 mg/kg. Notably, CHA maintained soil pH and SOM content, continuously reduced ACd content, and fixed RSF content under the rice-oilseed rape rotation system. CHA had a sustainable remediation effect on Cd-contaminated rice fields and was also cost-effective (1255.230 US $/hm^2). Based on Cd reduction efficiency, crop yield, soil environmental change, and total cost, CHA is recommended for sustainable remediation of Cd-contaminated acidic rice fields. These findings have significant implications for the safe production of grains and oil crops under high Cd contamination in karst mountainous areas.

Funding

This research was funded by the National Nature Science Foundation of China (42267038 and 42167003), the Science and Technology Project of Guizhou Province (Guizhou Science Foundation-Zhongke [2022] No. 054), and the Youth Science and Technology Talents Development Project of General Colleges and Universities in Guizhou (Guizhou Provincial Department of Education, Qianjiaohe Qizi [2022] No. 154).

CRediT authorship contribution statement

Fei Lou and Tianling Fu: Conceptualization, Methodology, Visualization, Writing, and editing. **Guandi He, Weijun Tian, and Jichang Wen** performed the field experiments, data analysis, and editing. **Mingfang Yang, Xiaoliao Wei, and Yeqing He:** Methodology. **Fei Lou and Tengbing He:** Conceptualization, Editing, Funding acquisition. All authors have read and agree to the published version of the final manuscript.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgement

We thank all colleagues in the research group for their support and assistance in conducting relevant experiments. We are also grateful to the chief editor, editor, and reviewer for objectively assessing our

manuscript and for providing constructive comments and suggestions to further improve the manuscript content.

Author Contributions Statement

All the authors agreed to the content of the manuscript and its submission to the journal, and all the authors listed have made significant contributions to this work.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2023.114884.

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