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ORIGINAL ARTICLE

Effects of water–organic fertilizer coupling on immobilization remediation technology using sepiolite

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Abstract
Sepiolite shows great potential to reduce pollution risk of cadmium (Cd)-contaminated rice of acidic paddy soils, but its effect was the synergy of water–organic fertilizer coupling in practical application. In this study, we studied the effect of goat manure (GM) in primordial and sepiolite (SP) amended soil under aerobic, intermittent, and flooded irrigation condition. The results showed that the application of goat manure in SP-amended soil increased soil pH by 15.6%–23.6%. The application of goat manure increased the content of DOM by 244.3%, 135.2% in unamended and sepiolite amended soil, respectively. As a consequence, the application of GM increased available Cd extracted by DTPA in SP-amended soil under aerobic and flooded condition, but decreased available Cd under intermittent irrigation condition. The application of sepiolite alone in Cd polluted soil decreased the accumulation of Cd in unpolished rice at the first year, but Cd content witnessed slight increase at the second year. Under intermittent condition, the application of goat manure in SP-amended soil decreased Cd content by 229.2%, 59.3% of unpolished rice in the first and second year, respectively. As a result, the application of goat manure and intermittent irrigation pattern could present a water–organic fertilizer coupling effect in SP-amended soil and further increased the long-term passivation effect of sepiolite.

Keywords Cadmium · Goat manure · Sepiolite · Rice · Water management

Introduction

Soil is the main gathering place for metals and acts as a barrier to prevent them from transferring through food chain to protect food safety and human health. It is reported that around 2.35×10^8 hectares farmland are contaminated with trace metal elements worldwide (Bermudez et al. 2012). In China, due to improper disposal of industrial wastes and the excessive use of pesticides, fertilizers and plastic films, pollution caused by heavy metal, such as Zn, Cd, Cr, Cu, and Ni, is getting serious and the pollution area is gradually expanding (Liang et al. 2016; Ekoa Bessa et al. 2020). According to a general survey of soil pollution status released in 2014, Cadmium (Cd) is the most widely distributed soil pollutant in China. In addition to natural processes (e.g., volcanic eruptions, rock weathering, erosion, atmospheric deposition, conversely and deposition of dust), human activities are changing soil functions and properties more rapidly and more intensively. More than 90% of Cd release in the environment is caused by man-made sources (Bi et al. 2006). Therefore, the prevention and control of Cd pollution has become a hotspot in the field of soil environment.

Engineered soil remediation technology (such as mixing of soil method) and bioremediation technology (such as phytoremediation and microbial remediation) are difficult to realize in practical applications due to high cost and low remediation efficiency. Therefore, as soon as the in situ chemical immobilization remediation technology appeared, it has attracted widespread attention for its advantages of high cost-effectiveness, easy to operate, suitable for large-area soil treatment, and environmental friendliness.

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Immobilization remediation is a process of converting metals with high mobility into solid or physicochemical stable form through chemical interactions among heavy metals, soil particles, and binders while agricultural production (He et al. 2020). Numerous soil amendments, i.e., alkaline compounds (Liu et al. 2020), clay minerals (Xie et al. 2018), metal oxides (Lin et al. 2021), organic scraps (Jin et al. 2020) and phosphate sulfides (Song et al. 2021) have been proposed and evaluated for their ability to immobilize heavy metals in soil environment. The mechanism involved in Cd adsorption of natural clay mineral containing redox potential (Fulda et al. 2013), soil pH (Yin et al. 2017), ion-exchange (Bolan et al. 2013; Luo et al. 2013), surface complexation (Liang et al. 2017) and precipitation reactions (Khaokaew et al. 2011; Agrawal and Sahu 2006).

Therefore, in the past 2 decades, the research on heavy metal pollution using immobilization remediation method has mainly focused on the screening and synthesis of immobilizing agents. Our previous (Li et al. 2018; Liu et al. 2019a, b; Sun et al. 2021; Yang et al. 2021; Wang et al. 2021a, b) study found that the application of natural sepiolite could decrease available content of Cd in soil and reduce the pollution risk of Cd on plant. However, natural sepiolite might not achieve the desired effect during planting and other soil biogeochemical process, like acid-soluble and complexation effects. As a consequence, development of agronomic management technologies to prevent Cd remobilization was an important pathway to control the food safety of agricultural products in soils.

As we all know, continuous flooding irrigation dramatically reduced Cd accumulation in rice grains (Wiggenhauser et al. 2020). Because, radial oxygen loss from rice roots temporarily oxygenates the rhizosphere and causes redox oscillations in the soil (Maisch et al. 2019). Rinklebe et al. (2016) and Ye et al. (2018) found that the reduction of sulfate to sulfide and the formation of insoluble CdS consequently reduced available Cd in the flooded soil. Intermittent flooding promote rice growth through regulate of soil permeability. The periodic variation in rhizosphere soil may affect the dominant biogeochemical reactions of redox-sensitive elements such as Fe, nitrogen (N), sulfur (S) and carbon (C) (Zhu et al. 2014). Meanwhile, intermittent irrigation improves rainfall utilization rate and facilitate water conservation (Lv et al. 2014).

As the main source of plant nutrition, the application of chemical and organic fertilizer also affects the distribution of soil heavy metal fractions (Li et al. 2016). Wang et al. (2021a, b) found that ammonia (NH_4^+-N) accelerated the remobilization of immobilized Cd. However, the impact of organic manure (OM) on mobility and phytotoxicity of heavy metal remains controversial. On one hand, OM reduces available Cd in the process of complexation or chelation (Ren et al. 2016; Mohamed et al. 2010). OM influences the Cd translocation into plants during soil nutrient uptake, root exudates, rice transpiration and transport protein activity (Rizwan et al. 2015). In contrast, OM may induce metal mobilization through the formation of small molecular organic acids (Richard et al. 2009; Salati et al. 2010).

We had already known that continuous flooding irrigation inhabitant the accumulation of trace metal in rice grains. But, the application of organic manure in soil amendment treatments are still questionable on account of type of fertilizer, especially fertilizers with different acids and alkalinity. Furthermore, water and fertilizer coupling effect is unavoidable and inevitable affecting factors in stabilization remediation treatment. Accordingly, the objectives of this study were to investigate the effects of common water management and goat manure on SP-amended paddy soil.

Materials and methods

Physicochemical properties of sepiolite, goat manure and soil

Soil specimen with natural Cd was collected from the 0 to 20 cm layer in Chenzhou, Hunan, China (112°43′48"N, 25°43′48"E). The basic characteristics of soil are shown in Table 1. Natural sepiolite was purchased from Huakai Environmental Protection Materials Co., Ltd in Hebei. The composition found to be 41.7% CaO, 16.8% MgO, 7.4% Al_2O_3, and 32.5% SiO_2 with the main minerals being $CaCO_3$, sepiolite, and SiO_2. No Cd was detected in natural sepiolite samples. The goat manure (GM) was purchased from a local fertilizer market in Tianjin, China. Selected characteristics of goat manure are listed in Table 2.

Experimental setup and water management

Pot experiment in the first year

Location and climate Pot experiments were performed in a greenhouse in the Agro-Environmental Protection Institute, Tianjin, China (117°9′35"N, 25°6′17"E). This area is char-

Table 1 Basic characteristics of soil

Properties	Value
pH (H_2O)	6.21
OM ($g·kg^{-1}$)	34.6
Total N ($mg·kg^{-1}$)	42.9
Available P ($mg·kg^{-1}$)	9.3
Available K ($mg·kg^{-1}$)	182.7
CEC ($cmol·kg^{-1}$)	25.3
Total Cd ($mg·kg^{-1}$)	2.5

Table 2 Basic Characteristics of goat manure

Properties	Value
pH	10.1
OM(%)	92.1
Total N (%)	2.54
Total P (%)	0.56
Total K (%)	1.09
Total S (%)	0.56
Total Cd ($mg \cdot kg^{-1}$)	0.74

Table 3 Summary of pot experiment treatments

	Amendment	Amount of sepiolite	Amount of goat manure	
			0	1.0% (*w/w*)
Flooded	Goat manure	–	CK	GM
	Sepiolite (SP) + goat manure (GM)	1.0%	SP	SG
Intermittent irrigation	Goat manure	–	CK	GM
	Sepiolite (SP) + goat manure (GM)	1.0%	SP	SG
Aerobic	Goat manure	–	CK	GM
	Sepiolite (SP) + goat manure (GM)	1.0%	SP	SG

acterized as a warm temperate and semi–humid continental monsoon climate, with the mean annual temperature is 13.7℃ and a frost-free period of 246 days.

Experiment design The soil samples was air-dried, homogenized and ground to pass through a 4 mm mesh sieve. A 6.0 kg soil samples containing 3.87 g urea and 6.24 g monopotassium phosphate was thoroughly mixed into each plastic pot (23.3 cm in diameter, 34 cm in height) (Xiao et al. 2015). Treatments containing base fertilizers alone were set as blanks. The treatment comprised sepiolite and goat manure are shown in Table 3. Each treatment was performed in triplicate.

Soil culture Each pot was equilibrated for 1 month, maintaining approximately 75%of field water-holding capacity (WHC) with tap water (without Cd). In order to determine soil moisture content, the pots with a certain amount of soil were submersed in water for 24 h following a subsequent drainage period of 24 h while preventing evaporation. The recorded weight of the water held in the soil after drainage was taken as WHC (Hansen et al. 2016).

Planting scheme Seedlings of rice (*Oryza sativa* L.) were germinated on perlite and transplanted on June 11, 2019. The rice plants were grown under flooded conditions for 21 d before adopt different water treatment. In the growing season, there were a total of three water treatment conditions, involving continuous flooded (CF, maintained 2–3 cm water layer in pot throughout the growth period), intermittent irrigation condition (IW, repeated every 7-d with 3-d flooding followed by 4-d drainage), and aerobic condition (AO, soil maintained moisture during the growth period). Drainage from the pots was prevented during the experiments. The pots were arranged in random blocks and rotated intermittently to ensure same conditions for each pot.

Pot experiment in the second year

To test and verify the long-term stability and effectiveness of sepiolite, pot experiment with the same design was conducted in the second year. The pots have applied sepiolite in the first year, but longer supplement in the second year. The pot have applied goat manure in the first year were added with the same dosage of GM to replenish soil fertility.

Sample preparation and analysis

After 135 d of growth, the plants were harvested and separated into root, straw, rachis, husk, and rice grains. Each part of the plant were ground and passed a 1.85 mm sieve for further analysis. The rice grains were oven-dried at 75 °C and weighed immediately after removing the husk and unfilled grains. Cd contents in plant samples were digested using a block digester and the supernate was determined by inductively coupled plasma mass spectrometry (iCAP Q; Thermo Scientific, Waltham, MA, USA) (Yin et al. 2017).

Rhizosphere soil was collected from a small amount of soil shaken directly from the plant roots. Soil samples were air-dried at room temperature and sieved to pass through 1 mm sieve, and stored in a plastic container for further analysis.

Soil pH was measured with a pH meter (PB-10, Sartorius, Germany) at the solid-to-liquid ratio (m:v) of 1:2.5 (Zhou et al. 2018a).

DOM content was measured using a total organic carbon analyzer (Vario TOC, Elementar Germany element) at a solid-to-water ratio of 1:5 (Gao et al. 2018).

The bioavailability of Cd in soil was determined by diethylenetriaminepentaacetic acid (DTPA) at a solid-to-water ratio of 1:5 (Liang et al. 2016; Wang et al. 2016a, b).

Changes in the fractions of Cd in paddy soil were analyzed by sequential extraction (Tessier et al. 1979), which separates heavy metals into five different fractions: extractable with 1.0 M $MgCl_2$ (pH = 7) (exchangeable, EXC-Cd), extractable with 1.0 M NaOAc (pH = 5) (bound to carbonates, CB-Cd), extractable with 0.04 M $NH_2OH·HCl$ in 25% (vol.%) HOAc solution (bound to iron and manganese oxides, OX-Cd), extractable with 0.02 M HNO_3 in 30% (wt.%) H_2O_2 (bound to organic matter, OM-Cd), and HNO_3–HF–$HClO_4$ digested (residual, Res-Cd).

Quality control

To monitor the accuracy and quality of chemical analyses, quality control measures were adopted using soil (GBW (E)-070,009) and plant reference materials (GBW-10045 (GSB-23)) obtained from the Institute of Geophysical and Geochemical Exploration (IGGE, China). Three reagent blanks and three standard reference materials were included in each batch of extraction, preparation, and analysis of every 40 samples. The recovery rate were 95%–103% and 92%–100% for Cd of plants and soil, respectively.

Statistical analyses

All treatments were replicated three times. The means and standard deviations (SD) were calculated using Microsoft Office Excel 2003. Two way analysis of variance was carried out with SAS 9.2 (SAS Inc., NC, USA). The mean values of the experimental parameters were compared using Student's *t* test at $p \leq 0.05$ (significant).

The ability of different part of rice to translocate and take up Cd was assessed using the translocation factor (TF_p). TF_1, Translocation factor from root to straw; TF_2, Translocation factor from straw to rachis; TF_3, Translocation factor from rachis to husk; TF_4, Translocation factor from husk to unpolished rice.

$$TF_1 = \frac{C_s}{C_r}$$

$$TF_2 = \frac{C_{ra}}{C_s}$$

$$TF_3 = \frac{C_h}{C_{ra}}$$

$$TF_4 = \frac{C_{up}}{C_h}$$

where C_r, C_s, C_{ra}, C_h, and C_{up} are the contents of Cd in roots, shoots, rachis, husk, and unpolished rice, respectively.

Results

Effects of GM on grain yield under different water management treatments

As shown in Fig. 1, grain yield was higher in intermittent irrigation condition, comparing to aerobic and flooded condition. In unamended soil, the application of GM increased grain yield by 20.6% and 30.1% in aerobic and intermittent condition, but decreased grain yield by 20.3% in flooded paddy soil. In sepiolite amended soil, the application of GM increased grain yield by 34.3% and 40.2% in aerobic and

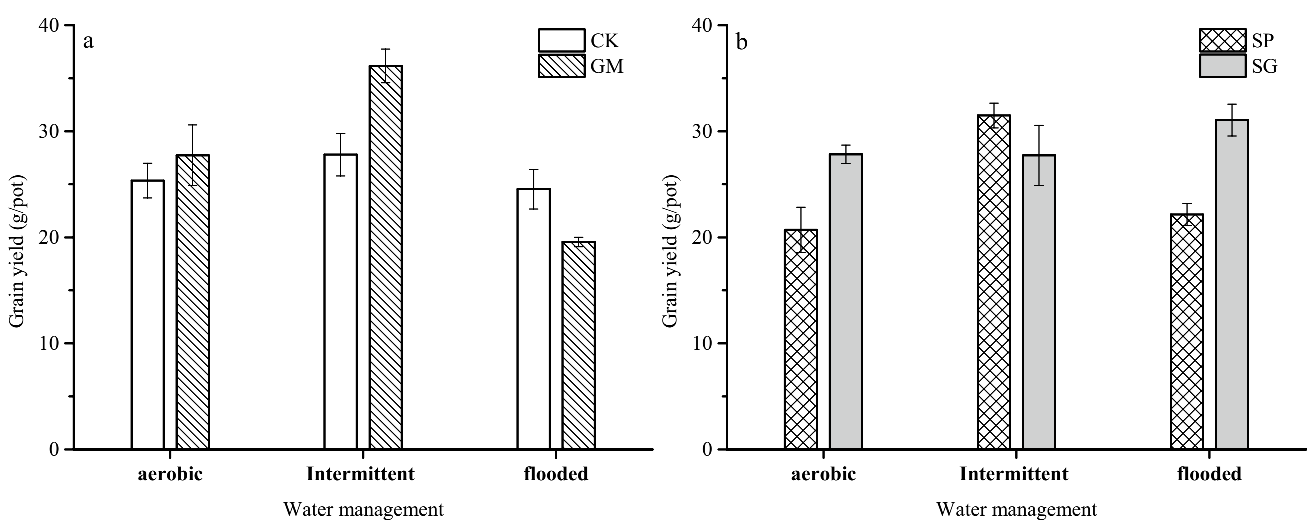

Fig. 1 Effects of goat manure on grain yield under different water management treatments (*CK* no material was applied, *GM* 1.0% (*w/w*, the same below) GM was applied to the pots; *SP* 1.0% SP was applied to the pots; *SG* 1.0% GM and SP was applied to the pots.)

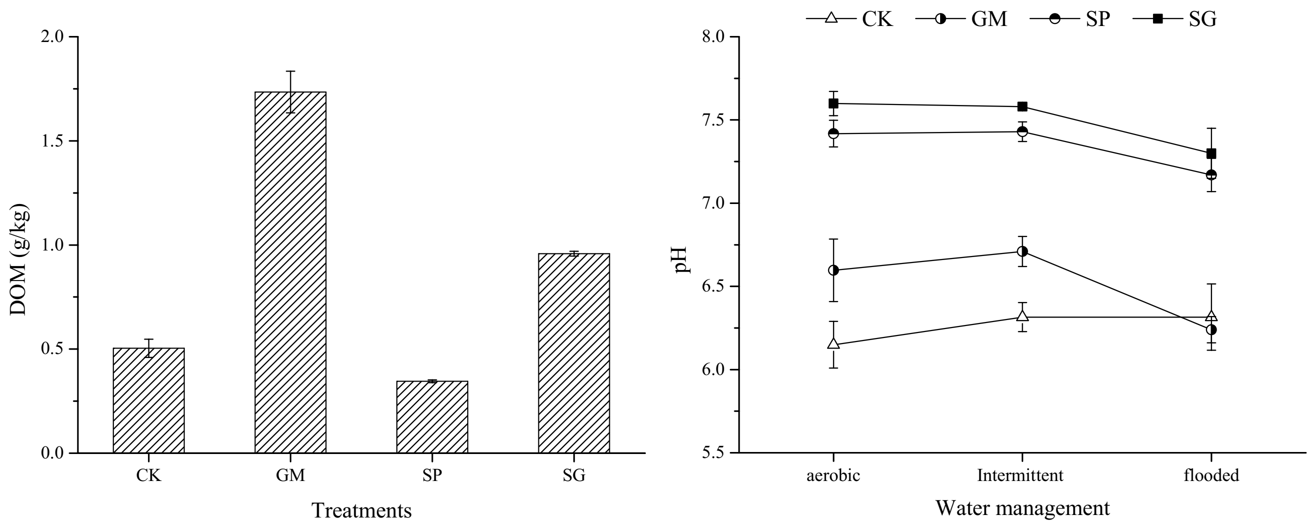

Fig. 2 Effects of goat manure on pH and DOM under different water management treatments

flooded condition, while decreased grain yield by 11.9% in intermittent irrigation.

Effects of GM on soil DOM and pH under different water management treatments

As shown in Fig. 2, in the control treatment, soil pH slightly increased in intermittent and flooded paddy soil. The application of sepiolite alone increased soil pH by 13.5%–20.6% under different water condition. The application of GM increased pH by 6.23%–7.27%, 1.81%–2.44% in unamended and SP-amended soil, respectively. The application of GM increased DOM by 244.3%, in contrast, the application of sepiolite decreased DOM by 31.5%. Therefore, the application of GM in SP amended soil increased DOM by 135.2%. Dissolved organic matters refers to the organic matter that can be extracted by water passing 0.45 μm membrane.

Effects of GM on Cd accumulation factor in different parts

As shown in Table 4, the translocation factors of different parts were directly related to the Cd accumulation character. The translocation factors (TF) from roots to straws were between 0.08 and 0. 24. The transfer factors from straws to rachis were between 0.31 and 0.55. The translocation factors from rachis to husk showed great disparity under different water management. The translocation factors from rachis to husk were between 0.31 and 0.67 under aerobic and intermittent irrigation condition, whilst the translocation factors from rachis to husk were 0.86–2.00 under flooded condition. As for the translocation factor from husk to unpolished rice, the only TF < 1 was obtained for sepiolite-amended soil with the application of goat manure under flooded and intermittent irrigation condition. The translocation factor from root to unpolished rice was the lowest when applied goat manure in SP-amended soil under intermittent irrigation condition.

Effects of GM on Cd concentration of unpolished rice in two consecutive years

Table 5 shows the effects of GM on Cd of unpolished rice under different water condition. During the first year, in control treatments, Cd concentration of unpolished rice were 0.2908 mg·kg^{-1} when applied base fertilizers alone under flooded water treatment, whilst, Cd concentration were 4.85 times and 6.88 times higher under aerobic and intermittent water condition. The application of SP alone decreased Cd concentration by 51.3%–76.9%. The application of GM on unamended soil decreased Cd concentration by 49.7%–73.1%. The application of GM on sepiolite-amended soil decreased Cd concentration by 61.0%, 62.5% under aerobic and intermittent condition, whilst increased Cd concentration under flooded condition. In the following year, Cd concentration of unpolished rice were 0.4868, 0.3944, and 0.2385 mg·kg^{-1} under aerobic, intermittent, and flooded water treatment, respectively. Sepiolite-amended treatment decreased Cd concentration by 33.6%–46.7%. GM treatment decreased Cd concentration by 10.0%–47.3%. GM treatment on sepiolite-amended soil decreased Cd concentration by 32.6%–58.5%.

Effects of GM on available Cd of soil extracted by DTPA

Figure 3 shows the available Cd in soil extracted by DTPA of different treatment. The application of GM in unamended

Table 4 Cd content in different part of rice plant (mg/kg) and translocation factor

Treatments		Cd_{Root}	TF_1	Cd_{Straw}	TF_2	Cd_{Rachis}	TF_3	Cd_{Husk}	TF_4	TF_p
Aerobic	CK	46.42 ± 2.30	0.19	8.86 ± 0.80	0.49	4.33 ± 0.29	0.39	1.70 ± 0.07	1.00	0.0366
	GM	8.67 ± 0.18	0.16	1.43 ± 0.16	0.31	0.44 ± 0.01	0.57	0.25 ± 0.02	1.83	0.052
	SP	16.16 ± 2.00	0.10	1.65 ± 0.36	0.53	0.88 ± 0.002	0.31	0.27 ± 0.008	2.85	0.047
	SG	6.42 ± 0.60	0.13	0.85 ± 0.11	0.35	0.30 ± 0.02	0.57	0.17 ± 0.02	1.68	0.044
Intermittent	CK	42.21 ± 5.86	0.24	10.37 ± 0.28	0.55	5.70 ± 0.25	0.39	2.20 ± 0.17	1.04	0.054
	GM	15.72 ± 2.79	0.16	2.55 ± 0.05	0.44	1.12 ± 0.20	0.49	0.55 ± 0.001	1.65	0.057
	SP	17.09 ± 1.40	0.11	1.83 ± 0.004	0.42	0.76 ± 0.15	0.33	0.25 ± 0.02	2.12	0.032
	SG	9.09 ± 0.33	0.09	0.86 ± 0.01	0.42	0.36 ± 0.07	0.67	0.24 ± 0.01	0.86	0.022
Flooded	CK	3.24 ± 0.56	0.12	0.38 ± 0.002	0.37	0.14 ± 0.01	0.86	0.12 ± 0.001	2.42	0.092
	GM	2.06 ± 0.09	0.10	0.21 ± 0.02	0.48	0.10 ± 0.01	2.00	0.20 ± 0.005	0.73	0.070
	SP	4.54 ± 0.20	0.08	0.37 ± 0.05	0.43	0.16 ± 0.00	0.87	0.14 ± 0.01	1.01	0.030
	SG	2.12 ± 0.09	0.11	0.24 ± 0.01	0.42	0.10 ± 0.02	1.80	0.18 ± 0.002	0.95	0.079

TF_1 Translocation factor from root to straw; TF_2 Translocation factor from straw to rachis; TF_3 Translocation factor from rachis to husk; TF_4 Translocation factor from husk to unpolished rice, TF_p product of TF_1, TF_2, TF_3 and TF_4

Table 5 Effects of GM on Cd of unpolished rice

Treatments	Cd concentration($mg \cdot kg^{-1}$)			
	CK	GM	SP	SG
1st year				
Aerobic	1.6999 ± 0.0649	0.4570 ± 0.0001	0.7700 ± 0.0308	0.2866 ± 0.0116
Intermittent	2.2920 ± 0.1706	0.9089 ± 0.0711	0.5290 ± 0.0417	0.2062 ± 0.0190
Flooded	0.2908 ± 0.0008	0.1464 ± 0.0020	0.1415 ± 0.0077	0.1702 ± 0.0113
2nd year				
Aerobic	0.4868 ± 0.0418	0.2596 ± 0.0139	0.2566 ± 0.0105	0.1729 ± 0.0074
Intermittent	0.3944 ± 0.0328	0.2234 ± 0.0155	0.2885 ± 0.0157	0.1605 ± 0.0088
Flooded	0.2385 ± 0.0093	0.1583 ± 0.0101	0.2146 ± 0.0115	0.0891 ± 0.0061

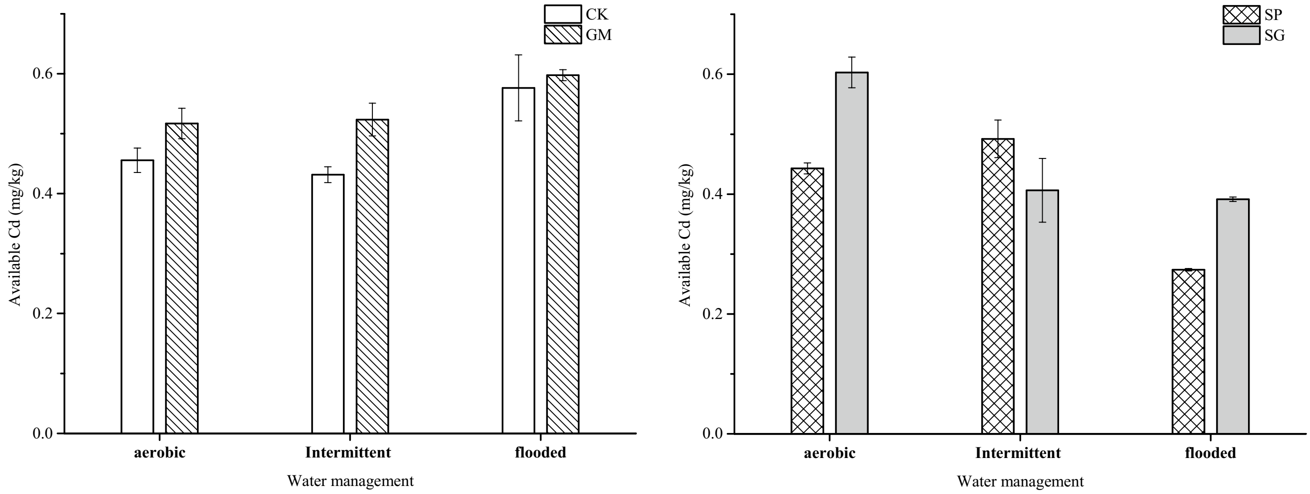

Fig. 3 Available Cd extracted by DTPA

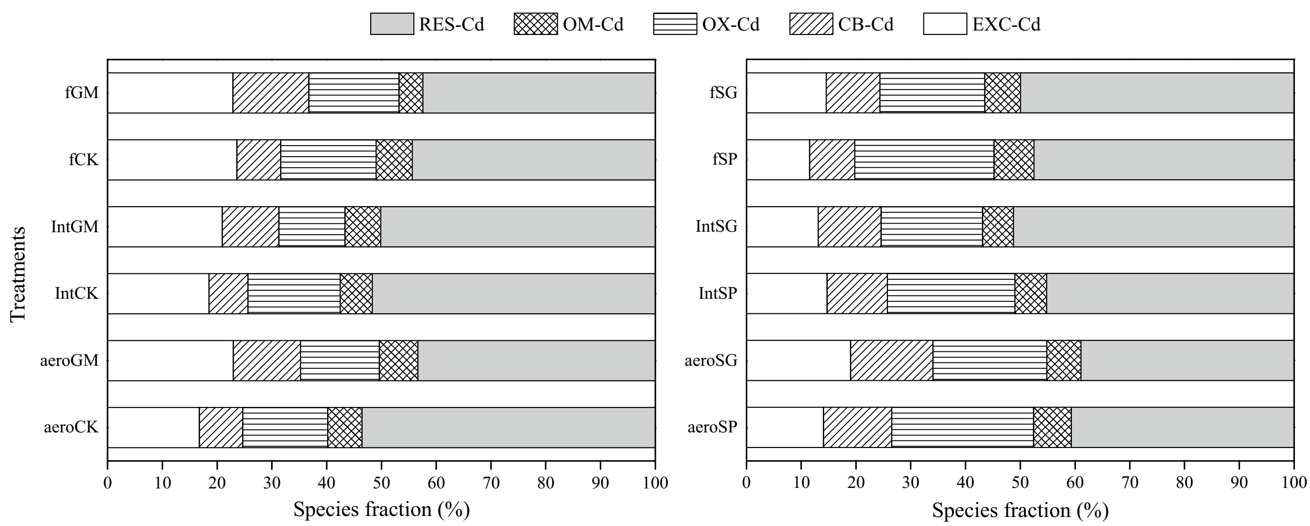

Fig. 4 Cd fractions of Tessier sequential extraction (*EXC-Cd* proportion of exchangeable Cd, *CB-Cd* proportion of Cd in carbonate-bound, *OX-Cd* proportion of Cd bounded with Fe–Mn oxides, *OM-Cd* proportion of Cd complexed with organic matter, *RES-Cd* proportion of Cd in residual form

soil increased available Cd extracted by DTPA under different water management. The possible explanation was due to the high application rate of GM, which has strong adsorption capacity and alkaline pH. The application of GM on SP-amended soil showed different effect on available Cd. Under intermittent irrigation condition, the application of GM in SP-amended soil decreased available Cd by 17.5%. The reason was that there would introduce greater carbonate and hydroxides phases into the soil solution, enhancing the formation of Cd precipitates with carbonates and hydroxides in subsurface soil under intermittent irrigation condition. However, the application of GM in SP-amended soil increased available Cd under aerobic and flooded irrigation condition.

Effects of GM on species distribution of Cd

Sequentially extractable Cd fraction percentages in the soil affected by GM are shown in Fig. 4. In the bulk soil of CK group, residual fraction accounted for a large proportion of the total Cd concentration (44.4%, 51.7%, and 53.6%, respectively). The application of SP decreased exchangeable fraction, and increased Fe-Mn-oxides bounded fraction. The application of GM decreased the residual fraction, whilst increased carbonate bounded fraction in unamended soil. In SP-amended soil, the application of GM decreased exchangeable fraction, increased the residual fraction simultaneously under intermittent irrigation condition. Whilst, under aerobic and flooded condition, the application of GM increased exchangeable fraction.

Discussion

Yield is the chief element to indicate the growth and development of crops that are affected by excess concentration of noxious substance in soil. It is reported that Cd might promoted the growth of plants with the concentration below 5 mg·kg^{-1} (Tang et al. 2020). Besides, water management and the application of soil amendments are other instabilities and uncertainties that might affect grain yield and production. Lv et al. (2014) found that the intermittent irrigation increased grain yield by 7.55%–29.58%. Song et al. (2021) found that comparing to continuous flooding, grain yield was higher under alternate wetting and drying (intermittent) irrigation condition. Furthermore, the application of phosphorus fertilizer slightly increased grain yield. Coincidentally, we found the same tendency in our research. Grain yield were higher under intermittent water condition comparing with flooded and aerobic water condition. The application of GM increased grain yield in unamended and SP-amended soil. Lahori et al. (2017) found that tobacco biochar and zeolite significantly decreased the dry biomass yield of *Brassica campestris L.* due to the increase in the soil pH compared to the control. In our study, the application of SP has no significant effect on grain yield. It might ascribe to the substantial reduction of dissolved Cd in soil. Soil acidity is a major yield-limiting factor that adversely affects crop productivity and Cd accumulation (Nanda and Adriano 2014). Ran et al. (2019) reported that the combine application of sepiolite and organic fertilizer increased soil pH by 1.0–1.2. In our study, the application of sepiolite and goat manure increased soil pH by 1.2–1.5 under different water condition, meanwhile grain yield increased by 9.7%–26.6%. This finding indicated that the combined application of sepiolite and organic manure can be used as a suitable choice for remediating Cd contaminated soil.

Soil pH and soluble carbon are important factors that influence the movement and bioactivity of heavy metals (Jiang et al. 2012). Zhou et al. (2014) described a negative correlation between soil pH values and bioavailable concentrations of soil Cd. Because with increasing soil pH values, concentrations of OH^- increased, followed by the OH^- and heavy metals forming a precipitate of hydroxide. Sepiolite, a pH-regulating immobilizing agent, could fix some heavy metals when absorbed to ion exchange sites. In our study, the soil pH is close to neutral and remain unchanged in planting season, whilst, soluble organic carbon increased when applied goat manure in unamended and SP-amended soil. However, throughout the growing season, these two factors affect the chemical forms and bioactivity of Cd in different ways (Kögel-Knabner et al. 2010). Many researchers also suggests that soluble organic carbon is the most important factor in terms of exchangeable Cd with high bioactivity (Xiao et al. 2006). In our study, the application of goat manure significantly increased soil DOM in unamended and SP-amended soil. Soil DOM increased by 2.44 times while applied goat manure alone. Li et al.(2018) found that soluble carbon increased significantly by 15% ($p \leq 0.05$) under the farmyard manure treatments. Bian et al. (2018) found that the application of silkworm excrement organic fertilizer increased DOM by 124.6%. As a result of large decreases in biomass inputs and soil organic carbon (SOC) concentrations due to the long-term effects of metal pollution on soil microorganisms and vegetation, Ekaterina et al. (2005) found that SOM complexed greater diversity and bond intensity of functional groups in unpolluted soils than in polluted soils. Zhou et al. (2017) studied the effects of organic matter fraction on distribution of Cd in long-term polluted paddy soils. The results showed that SOM fractions combined higher Cd than other soil constituents.

The root is widely recognized as the most major connection between rice plant and soil environment. Compared to aerobic and intermittent irrigation, flooded irrigation decreased Cd content of root by 73.4%–93.0%. Meanwhile, the accumulation pattern of Cd in other parts of rice plant was similar to that in root. Cd content in different part of rice ranked in the following order root > straw > rachis > husk. Water management affects the bioavailability of cadmium (Cd) in the soil and hence their accumulation in rice and grain yields. Compared with flooding, both aerobic and intermittent irrigation enhanced Cd distribution in the root and reduced it in the straw and grain. Translocation factors (TFs) were also the most perceptual intuition of the concentration between different parts. A decrease of Cd concentrations in unpolished rice was relevant to TFs in all parts of the rice plant, but different organs had different TFs. With the addition of different amendment, the TFs of Cd from roots to straw, straw to rachis, rachis to husks, husks to unpolished rice, and roots to unpolished rice changed to different degrees (Table 4). However, TFs are inversely proportional to biomass of different plant parts. We found that TFs from roots to straw all lower than 0.20. Because, the biomass of straws were much higher than roots. Wang et al. (2020) found that the application of hydroxyapatite decreased TF of maize, besides the effect increased with the increase of application rate. Chen et al. (2021) found that the ability of Cd to be transferred to the grains was effectively inhibited as the values of TF_G were lower than those of TF_S (TF_G, Translocation factor from straw to grains; TF_S, Translocation factor from root to straw). In our study, the TFs from husks to unpolished rice exceeded 1.0 in most of the treatments. This results indicated that ability of husks to transport Cd was greater than that of other rice organs. Table 4 shows that under intermittent water condition, the combined application of sepiolite and goat manure inhibited the transfer of Cd from roots to unpolished rice to the greatest extent.

In the first year, the combined application of sepiolite and goat manure decreased Cd by 83.1%, but did not reach the recommended tolerable levels as proposed in the national food pollutants standards (GB2762-2012; 0.20 mg·kg^{-1}). However, under flooded irrigation condition, the application of sepiolite alone, goat manure alone, and the combination all reduced the rice contamination risk. Cd content all below 0.20 mg·kg^{-1} in the amended treatments. In the second year, the application of sepiolite alone decreased Cd content of unpolished rice by 66.7%, 45.5% under aerobic and intermittent irrigation condition compared to the first year. But, under flooded condition, the application of sepiolite alone increased Cd content of unpolished rice in the second year. The reason may be the sepiolite particles subjected to flooded treatments were significantly damaged. Similar findings have also been reported by Spokas et al. (2014) and Cang et al. (2020). The results indicated that cropping pattern affected has a negative effect on the long-term stability of the sepiolite. But, the combined application of sepiolite and goat manure further decreased Cd content of unpolished rice under flooded condition. Therefore, the application of goat manure enhanced the effect and long-term stability of sepiolite.

It is well known that bioavailable fraction of metal is more important for plant uptake than the total concentrations of heavy metals in the soil which might be due to the presence of different chemical forms of heavy metals (Rizwan et al. 2016). Bioavailable Cd is the fraction of total Cd in the interstitial water and soil particles that is readily available to the receptor organisms. In our study, the application of goat manure slightly increased available Cd extracted by DTPA under aerobic and intermittent irrigation condition, but the effect did not reach a significant level. The DTPA-extractable metal concentration in the 10–20 cm profiles is adapted to assess the phytoavailable proportion of Cd. Therefore, the uptake and transportation of Cd by rice plants was also greatly influenced by DTPA-Cd (Fellet et al. 2014). Wang et al. (2016a, b) reported that the application of biochar decreased DTPA-Cd by 14%–51%, as a result, Cd in edible part decreased by 46%–86%. In this study, the application of goat manure increased DTPA-Cd in most treatments. Because organic manure increased -COOH of soil particle, which can promote the mobility of Cd by forming complexes. Zhou et al. (2018b) found that rice straw (RS) increased phytoextraction efficiency of Cd, and RS has positive effects on plant Cd uptakes.

In our study, the application of sepiolite decreased exchangeable Cd and increased the fraction bound to iron and manganese under different water irrigation condition. It is well established that iron and manganese oxides exist as chelates, concretions, between particles, or simply as a coating on particles, these oxides are excellent scavengers for trace metals and are thermodynamically unstable under anoxic conditions. The reason is that $CaCO_3$ occupied a large proportion in sepiolite. Besides, It will not decrease the residual form of Cd but the increase of goat manure enhances the adsorption of Cd on the manure itself (exchangeable fraction). But in SP-amended soil, the application of goat manure decreased exchangeable Cd and increased residual form of Cd under intermittent irrigation condition. The results showed that intermittent irrigation regime was a safe and effective pattern for Cd remediation using sepiolite. Besides, the application of goat manure in SP-amended soil was also a recommended measure to promote crop growth and achieve safe production in Cd polluted soil.

Conclusion

1. The application of sepiolite alone increased soil pH, as a consequence, sepiolite decreased available Cd under flooded condition, but increased available Cd under aerobic and intermittent irrigation condition.
2. The application of goat manure alone increased DOM content in unamended and sepiolite amended soil. As a result, goat manure increased available Cd under aerobic and flooded water condition in sepiolite amended soil.
3. Under intermittent water condition, the combined application of sepiolite and goat manure inhibited the transfer of Cd from roots to unpolished rice to the greatest extent.
4. The application of sepiolite in Cd-polluted soil decreased the accumulation of Cd in unpolished rice at the first year, but Cd content witnessed slight increase at the second year. But, the application of goat manure in sepiolite amended soil further decreased Cd content in unpolished rice under intermittent irrigation condition. As a result, the application of goat manure and intermittent irrigation pattern could present a water–fertilizer coupling effect in sepiolite amended soil and further increased the long-term passivation effect of sepiolite.

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Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

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