The risks of sulfur addition on cadmium accumulation in paddy rice under different water-management conditions

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A R T I C L E   I N F O

A B S T R A C T

Recently, the application of sulfur (S) has been recommended to control the accumulation of cadmium (Cd) in rice in contaminated paddy soil. However, the effects of exogenous S on Cd transfer in paddy rice systems under different water-management practices have not been systematically investigated. Pot experiments were performed to monitor the composition of soil pore water and the Cd accumulation in iron plaque and rice tissue were compared under different S (0 and 200 mg/kg Na_2SO_4) and water (continuous and discontinuous flooding) treatments. Sulfur application significantly increased Cd concentrations in soil pore water under discontinuous flooding conditions, but slightly reduced them under continuous flooding. Moreover, the oxidation/reduction potential (Eh) was the most critical factor that affected the Cd levels. When the Eh exceeded −42.5 mV, S became the second critical factor, and excessive S application promoted Cd dissolution. In addition, S addition elevated the Cd levels in iron plaque and reduced the Cd transfer from the iron plaque to rice roots. In rice, S addition inhibited Cd transfer from the rice roots to the straw; thus, more Cd was stored in the rice roots. Nevertheless, additional S application increased the Cd content in the rice grains by 72% under discontinuous flooding, although this effect was mitigated by continued flooding. Under simulated practical water management conditions, S addition increased the risk of Cd contamination in rice, suggesting that S application should be reconsidered as a paddy fertilization strategy.

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Introduction

Cadmium (Cd), a highly toxic heavy metal with high mobility in soil, can accumulate in food chains and seriously impact food security (Zhang and Reynolds, 2019). In southern China, anthropogenic activity, such as mining, smelting, and phosphate fertilization, has caused serious Cd contamination in soil (Hu et al., 2016). Rice (*Oryza sativa* L.) is cultivated throughout this region and is a staple food in China; however, compared with other cereals, rice readily accumulates Cd, which poses enormous risks to food safety and

human health (Song et al., 2017). Consequently, numerous studies have focused on developing methods to reduce the availability of Cd in paddy soil and Cd accumulation in rice grains.

Optimised water management and fertilization not only promote rice yields but also reduce the release of Cd in soil solutions, and thus potentially reduce its bioavailability. Previous studies have shown that increased waterlogging could effectively reduce Cd accumulation in rice plants (Zheng et al., 2019). Li and Xu (2017) found that compared with conventional irrigation, continuous flooding reduced the Cd content of brown rice by 39%, whereas wetting irrigation increased the Cd content by more than two times. Waterlogging leads to rapid soil oxygen depletion and organic matter (OM) decomposition by microorganisms, thereby decreasing the oxidation–reduction potential (Eh) and reducing oxides such as NO_3^-, Mn(IV), Fe(III), and SO_4^{2-} to N_2/NH_4^+, Mn(II), Fe(II), and H_2S. Free Cd^{2+} can form insoluble CdS with HS^- and S^{2-} or co-precipitates with FeS and ZnS, thus reducing Cd mobility (Feijtel et al., 1988). In addition, flooding-induced changes in soil pH can also affect the adsorption of Cd by colloids (Houben et al., 2013); however, the Cd activity in soil is dependent on complex soil environmental factors, for which the influencing mechanisms have not been clarified.

Recent studies have focused on the potential for sulfur (S) fertilization to suppress Cd transfer (Fan et al., 2010; Li et al., 2020; Wu et al., 2020; Zheng et al., 2019). In plants, S plays an important role in the synthesis and metabolism of proteins and nucleic acids (Stephan et al., 2013). Glutathione (GSH; γ-Glu-Cys-Gly) and phytochelatins (PCs; GSH oligomers) include sulfhydryl compounds that can protect plants against heavy metal stress (Fan et al., 2010; Li et al., 2020). Cao et al. (2018) found that an enhanced S supply increased the GSH and PCs contents in rice and reduced Cd accumulation in grains. Adhikari et al. (2018) found that supplemental sulfate in maize plants could maintain a high GSH pool for reducing redox stress conditions and alleviating Cd detoxification stress. Application of S reduced Cd absorption in the edible part of pakchoi owing to Cd chelation in the vacuoles (Li et al., 2020). In addition, an adequate S supply may reduce Cd accumulation by promoting Fe absorption by rice (Wu et al., 2020). In paddy soil, a waterlogging-induced anaerobic environment promotes the transformation of S-containing substances to S^{2-} and causes changes in the physical and chemical properties of the soil, including Eh and pH, thereby affecting the speciation, mobility, and bioavailability of Cd. Sulfur application can lead to greater S^{2-} production, and therefore Cd precipitation, under anaerobic soil conditions, thereby impeding Cd uptake by rice (Fulda et al., 2013; Yuan et al., 2019). Some researchers have also investigated the effects of different S forms on soil Cd. Liu et al. (2020) and Zhang et al. (2019) suggested that the Cd content of rice grains could be reduced more effectively by sulfate than by elemental S. Moreover, S addition under different water management strategies leads to various effects on Cd accumulation in rice (Liu et al., 2020; Zheng et al., 2019). Wu et al. (2019) revealed that S addition could enhance Cd accumulation in hyperaccumulators in non-flooded conditions but reduced the Cd content in rice grains under flooding conditions. Additionally, other studies have shown that S application can stimulate Cd

dissolution in soil under non-persistent flooding conditions (Li et al., 2019; Wang et al., 2020).

Based on previous findings, S is generally expected to reduce Cd bioavailability in soil and Cd accumulation in plants; however, such an effect may vary greatly or may even reverse under different water management conditions. During practical rice production, drying is usually conducted at the late tillering and filling stages to prevent ineffective tillering and protect root vitality to promote healthy rice growth. The specific anaerobic environments created by different water management practices alter S conversion and the effect of S on Cd availability in the soil. Therefore, it is necessary to investigate the effect of S on Cd accumulation in paddy soil under different water management practices by simulating practical cultivation water conditions and continuous flooding.

This study monitored Cd transfer and accumulation in a soil–rice system with the application of additional S under two different water-management approaches—(1) typical moisture conditions (drainage during late tillering and maturity and flooding throughout the rest of cultivation) and (2) extreme moisture conditions (continuous flooding)—to achieve a better understanding of the effects of exogenous S and different water-management practices on: (1) Cd mobility, by monitoring Cd, S, Fe, and Mn concentrations, and pH, Eh, and EC values of soil pore water; (2) the formation of and Cd absorption by iron plaque; and (3) the transfer and accumulation of Cd in rice plants.

1.   Materials and methods

1.1.   *Pot experiments*

Topsoil (soil from a depth of 0–20 cm) was collected from agricultural land in Wuzhou, Guangxi Province, China. The properties of the tested soil sample were as follows: 19% clay (< 0.002 mm), pH of 6.05, soil organic carbon of 20.02 g/kg, cation exchange capacity of 7.80 cmol/kg, total Cd of 0.15 mg/kg, total Fe of 29.97 g/kg, total Mn of 0.31 g/kg, total S of 327.19 mg/kg, and available S of 24.16 mg/kg. The tested soil was air-dried, passed through a 2 mm sieve, and dosed with soluble Cd (as $Cd(NO_3)_2$) in soil. After Cd addition, the soil was thoroughly mixed and aged in pots at a 60%−70% maximum water-retention capacity for one year. The Cd concentration in the aged soil was 1.52 mg/kg. After ageing, the soil was air-dried, passed through a 2 mm sieve, and mixed with a basal fertiliser (150 mg N/kg as $CO(NH_2)_2$, 30 mg P/kg as $Ca(H_2PO_4)_2 \cdot H_2O$, and 75.5 mg K/kg as KCl).

Two sets of experimental factors were considered, including two S application rates (0 and 200 mg/kg Na_2SO_4) and two water management regimes: continuous flooding without additional S (CF−S), continuous flooding with additional S (CF+S), discontinuous flooding without additional S (DF−S), and discontinuous flooding with additional S (DF+S). For the continuous-flooding treatments, the water surface was maintained at 3–5 cm above the soil throughout the growth period. For the discontinuous flooding treatments, soil moisture was maintained at 60%–70% of the soil water-retention capacity during the drainage periods in the late tillering and grain fill-

ing stages. Each pot contained 4.0 kg of soil and each treatment had three replicates.

Rice seeds (*Oryza sativa* L., Jingliangyouhuazhan) were sterilised with 30% H_2O_2 for 20 min and germinated on a moist net for 14 days. Two uniform rice seedlings and rhizosphere bags (15 cm height × 8 cm diameter; 40 μm) were transplanted into each pot on 2 May 2019. When the rice was moved into the rhizosphere bag, two soil pore water sampling devices (length: 10 cm, external diameter: 2.5 mm; Rhizosphere Research Products, Netherlands) were inserted into the soil at the same time. One sampling device was placed close to the rice root and the other was placed in the bulk soil at a distance of ∼10 cm from the root. Pot experiments were conducted at the China Agricultural University in a greenhouse maintained at 25±5 °C/20±2 °C, (day/night) under a 14 hr/day photoperiod with a homogeneous light intensity (240–350 mmol/m^2 s) and 70% relative humidity. The date of rice heading was 8 August 2019, which was 99 days after transplanting.

1.2. Sampling and ascorbic-citrate-acetic extraction of iron plaque

Throughout the rice-growing period, approximately 20 mL of rhizosphere soil pore water and bulk soil were collected simultaneously from each pot at 5, 12, 25, 50, 75, 120, and 158 days for continuous flooding treatments and at 5, 12, 25, 27, 32, 38, 75, 120, 122, 133, 143, and 158 days for discontinuous flooding treatments, using the soil pore water sampling devices and a syringe (50 mL). The 20 mL of pore water collected at each sampling time was divided into two samples. One part of the water sample (precisely 5 mL) was acidified using 5 mL of HNO_3 (5%, *V/V*) and stored at 4 °C until further elemental analyses. The other part of the sample (approximately 15 mL) was immediately analysed to measure the Eh, pH, and electrical conductance (EC) using an oxidation–reduction potential detector (ORP, HI 98,120, Hanna Instruments, Italy), a pH electrode (HI 98,127, Hanna Instruments, Italy), and an electrical conductance electrode (HI 8733, Hanna Instruments, Italy), respectively.

Plant samples were collected on the 160th day. Two mature rice plants from each pot were removed and assessed as one sample. After thoroughly washing the plants with deionised water, the mature rice plants were divided into roots, straw, husk, and grains. The iron plaque on the root surfaces was then extracted using ascorbic-citrate-acetic (ACA; 0.3 mol/L sodium citrate, 10% sodium acetic (*V/V*), and 3 g ascorbic acid; Zou et al., 2018). The fresh roots were immersed in 90 mL of ACA and shaken at 280 r/min for 3 hr at 25 °C. The resulting solution was accurately diluted to 200 mL with deionised water and filtered through quantitative filter papers (0.45 μm). The filtered samples were stored at 4 °C until analyses. Samples of each part of the rice plant were further dried at 75 °C for 48 hr, weighed to calculate their biomass, and then completely ground.

1.3. Determination of Cd, Fe, Mn, and S contents

To measure the total heavy metals and metalloids in rice plants, 0.25 g of dried plant material was microwave-digested (Mars 5; CEM, USA) with 8 mL of HNO_3 (65%; *V/V*) at 180 °C

for 40 min, after which the acid was removed. The digestion solution was stabilised to 50 mL with ultrapure deionised water and filtered through a 0.45 μm filter. The Cd concentration of the digestion solution was determined via inductively coupled plasma mass spectrometry (ICP-MS 7700, Agilent Technologies, USA), and the Fe, Mn, and S concentrations were determined via inductively coupled plasma optical emission spectroscopy (ICP-OES OPTIMA 7300 DV, Perkin-Elmer, USA). The Cd concentrations of the acidified soil pore water samples and ACA extractions were measured using ICP-MS, and the Fe, Mn, and S concentrations were determined using ICP-OES. For quality assurance, two reagent blanks and a standard reference material (GSB-26, celery; National Institute of Metrology, China) were included in each batch. The recovery rates for the reference material ranged from 87% to 115%.

1.4. Data analysis

The results were uniformly analysed based on dry weights, and data were presented as mean ± standard error (SE). The analysis of variance results was calculated using SAS 9.0 (least significant difference, LSD; P < 0.05). The various analyses were performed as follows:

Translocation factors (TF) were calculated as the ratio of Cd content (*C*) in different sample parts:

$$\text{TF}_{\text{root/ironplaque}} = \frac{C_{\text{root}}}{C_{\text{iron plaque}}};$$

$$\text{TF}_{\text{aerialpart/root}} = \frac{C_{\text{aerial part}}}{C_{\text{root}}};$$

$$\text{TF}_{\text{straw/root}} = \frac{C_{\text{straw}}}{C_{\text{root}}};$$

$$\text{TF}_{\text{husk/straw}} = \frac{C_{\text{husk}}}{C_{\text{straw}}};$$

$$\text{TF}_{\text{grain/husk}} = \frac{C_{\text{grain}}}{C_{\text{husk}}}. \tag{1}$$

The total Cd content (T_{Cd}, μg) was determined as follows:

$$\begin{aligned} T_{\text{Cd}} = {} & C_{\text{iron plaque}} \times W_{\text{iron plaque}} + C_{\text{root}} \\ & \times W_{\text{root}} + C_{\text{aerial part}} \times W_{\text{aerial part}} \end{aligned} \tag{2}$$

where *C* (mg/kg) is the content of Cd and *W* (g) is the dry weight.

The Cd distribution rate (*D*; %) within iron plaque, roots, and aerial parts was calculated as follows:

$$\begin{aligned} D_{\text{ironplaque}} &= \frac{C_{\text{iron plaque}} \times W_{\text{iron plaque}}}{T_{\text{Cd}}} \times 100\%, \\ D_{\text{root}} &= \frac{C_{\text{root}} \times W_{\text{root}}}{T_{\text{Cd}}} \times 100\%, \\ D_{\text{aerialpart}} &= \frac{C_{\text{aerial part}} \times W_{\text{aerial part}}}{T_{\text{Cd}}} \times 100\%. \end{aligned} \tag{3}$$

Conditional inference trees (CIT) are a popular tool for regression analysis to categorise data based on binary recursive partitioning in a conditional inference framework (Zheng et al., 2020). The relationships between the Cd concentrations in the soil solutions, the concentrations of Fe, Mn, or S, and soil properties (pH, Eh, or EC) were analysed using CIT to identify the key factors that impacted Cd concentrations in

soil pore water based on significance testing. The CIT analysis selected crucial factors that affect Cd dissolution by significance testing, and then classified the dataset into several categories based on the significance levels. The specific steps were as follows: (1) calculate the P values between the Cd concentration (output variable) and other properties (Fe, Mn, and S concentrations; pH, Eh, and EC values; predictor variables); (2) conduct binary recursive partitioning between the dependent variable and the selected variable with the minimum P value, then select the most significant segmentation; and (3) categorise the dataset into two parts, repeating the previous steps in the subset until the smallest node is implemented (Hothorn et al., 2006). The CIT and correlations were calculated using the R 3.6.2 software package.

2. Results

2.1. Elemental and soil pore water property dynamics

The soil pore water Eh and pH were significantly influenced by different water conditions. Under DF, the Eh decreased when the soil was flooded and increased when the soil was exposed to air (Fig. 1a). Under CF, after 12 days of flooding, the Eh dropped to approximately −100 mV and remained stable thereafter. The average Eh values from the 12th through 158th day of rice cultivation (DRC 12–158) were −112 mV (rhizosphere) and −107 mV (non-rhizosphere) without additional S and −92 mV (rhizosphere) and −82 mV (non-rhizosphere) with additional S. Thus, S addition increased the Eh value, but only under CF (Fig. 1b). Moreover, the effect of water conditions on pH differed from that on Eh. Under both CF and DF, flooding increased the pH of the soil pore water. In the early stage of rice cultivation under both water conditions, the pH of the pore water with additional S was lower than that without additional S, whereas in the middle and late stages of rice cultivation, additional S slightly increased the pore water pH (Fig. 1c, d). Regardless of water management strategy and S application conditions, the pH of rhizosphere soil was lower than that of non-rhizosphere soil (Fig. 1c, d). Additionally, the EC value increased significantly owing to the added S. Throughout the cultivation process, the EC exhibited a downward trend, though it clearly increased when the water was initially drained under DF conditions. In the middle and late stages of rice cultivation, the EC of rhizosphere soil was greater than that of non-rhizosphere soil (Fig. Appendix A S1c, d).

Sulfur addition significantly increased the soil pore water S concentration under all treatments. For both water treatments, the first samples under all treatments on DRC 5 showed that S addition increased the S concentrations in the pore water by more than five times. Under DF, the S concentrations decreased rapidly when the soil was first flooded and increased sharply after drainage. However, S dissolution was much less pronounced after the second drainage (Fig. 1e). Under CF, regardless of S addition, S concentrations decreased sharply in the first 75 days and then remained below 6 mg/L. S concentrations were less than 3 mg/L at the end of cultivation (DRC 158; Fig. 1f). The Fe and Mn concentrations in the soil pore water increased by flooding and decreased by drainage,

though the effect on Fe was more evident. Under DF, the Fe and Mn concentrations reduced to less than 1.83 and 0.72 mg/L, respectively, after the second drainage. Under both water management strategies, S addition promoted Fe and Mn dissolution in the early stage of cultivation but inhibited dissolution in the middle and late stages. This phenomenon was more pronounced under CF, and the effect on Fe dissolution was greater than that on Mn dissolution. In the middle and late stages of the CF treatments (DRC 75–158), S addition decreased the Fe concentrations by 23%–57%, although Fe and Mn concentrations in rhizosphere soil were higher than those in non-rhizosphere soil during this period (Appendix A Fig. S1e–h).

In general, drainage promoted the dissolution of Cd in soil pore water, whereas flooding reduced it. Under DF, the Cd concentrations in soil pore water decreased by more than 95% within 25 days of flooding (DRC 5–25) under both S treatments. Subsequently, after 13 days of drainage (DRC 38), the Cd concentrations increased to peak values of 5.49 μg/L (rhizosphere) and 5.34 μg/L (non-rhizosphere) without additional S and 8.69 μg/L (rhizosphere) and 7.35 μg/L (non-rhizosphere) with additional S. Subsequent re-flooding drastically decreased the Cd concentrations by at least 82%. However, subsequent re-drainage did not significantly affect Cd dissolution (Fig. 1g). Under CF, the Cd concentrations rapidly decreased below 3 μg/L within 12 days of flooding for both S treatments and remained stable at < 0.7 μg/L until the end of rice incubation (DRC 23–158; Fig. 1h).

Notably, the effect of S on Cd dissolution varied under different water management conditions. Under DF, additional S consistently increased the Cd concentrations in the soil pore water by 18%–126% over those in soil pore water without additional S during DRC 32–158 (Fig. 1g). Under CF, the Cd concentration in the rhizosphere pore water with S addition reduced by 21%–83% compared with that without additional S during DRC 23–158 (Fig. 1h). However, under both water management conditions, the Cd concentrations of the rhizosphere soil pore water were higher than those in the bulk soil during the middle and late stages of rice cultivation (Fig. 1g, h).

2.2. Correlation analysis of key soil pore water characteristics

The Cd concentration of the soil pore water was significantly negatively correlated with Fe concentration ($r = -0.34$, $P < 0.05$) and pH ($r = -0.24$, $P < 0.05$), whereas it was significantly positively correlated with Eh ($r = 0.47$, $P < 0.05$), EC ($r = 0.40$, $P < 0.05$), and S concentration ($r = 0.32$, $P < 0.05$). No significant correlation was observed between Cd and Mn concentrations. The S concentrations of the soil pore water were significantly positively correlated with Fe and Mn concentrations and EC values, but negatively correlated with pH (Fig. 2a).

To identify the key factor (i.e., the factor that most significantly affected Cd dissolution in paddy soil), the CIT method was introduced based on significance tests. The CIT method can dissect the logical relationships between crucial factors. The Cd concentrations under different influencing factors were shown in the form of a box diagram. The CIT results indicate that Eh and S concentrations were the critical factors influencing the Cd concentrations of soil pore water (Fig. 2b). According to the first fork point, Eh was generally the most
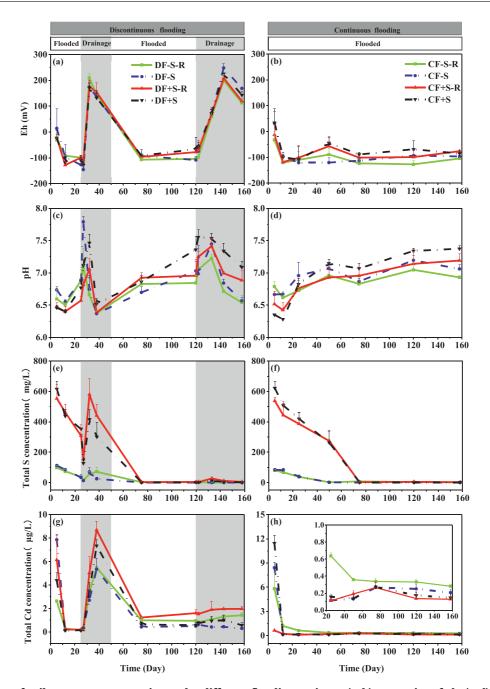
Fig. 1. – Behaviour of soil pore water properties under different flooding regimes. (a, b) Dynamics of Eh, (c, d) pH, (e, f) total S, and (g, h) total Cd concentrations in soil pore water under discontinuous flooding (a, c, e, g) and continuous flooding (b, d, f, h) conditions. −S-R: rhizosphere soil without additional S; −S: bulk soil without additional S; +S-R: rhizosphere soil with additional S; and +S: bulk soil with additional S. Data is presented as mean ± standard error (SE), where $n = 3$.

critical factor controlling Cd dissolution. When Eh was less than −42.5 mV, the average Cd concentration remained at a relatively low level of 0.48 μg/L (mean values are shown in the first box diagram of the CIT chart), and the S concentration did not significantly affect Cd concentrations in the soil pore water. However, when the Eh value exceeded −42.5 mV, S concentrations became the second critical factor affecting the Cd concentrations of soil pore water. Compared with the average Cd concentration (0.48 μg/L) at an Eh less than −42.5 mV, the average Cd concentration (1.40 μg/L) at an Eh over −42.5 mV

increased by approximately three times for S concentrations less than 51.3 mg/L, while the Cd concentration (6.27 μg/L) for S concentrations greater than 51.3 mg/L increased by 13 times (Fig. 2b).

2.3.　Cd in iron plaque

A layer of reddish-brown attachments could be observed on the surface of the rice roots and colour was deeper red under CF than under DF treatments. The CF treatments increased
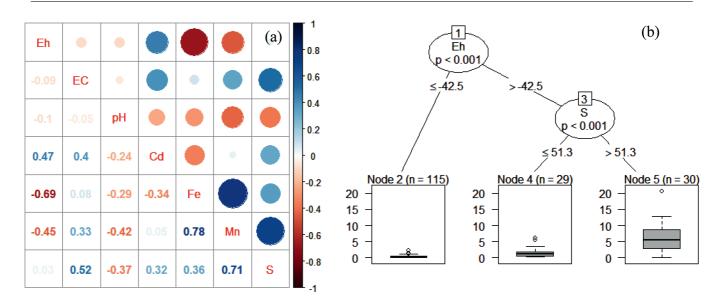
Fig. 2. – Statistical analysis of soil pore water properties. (a) Correlation analysis of Cd, Fe, Mn, and S concentrations with Eh, EC, and pH values of soil pore water; (b) conditional inference tree (CIT) for factors that influence Cd concentrations, where box plots represent the Cd concentrations (μg/L) in soil pore water under different categories and the black line in box plots indicates the mean value. The units of Eh and S are mV and mg/L, respectively.

Table 1. – Fe, Mn, S, and Cd content in iron plaque on the surface of rice roots.

Treatment	Fe (g/kg)	Mn (g/kg)	S (g/kg)	Cd (mg/kg)
DF−S	14.38 ± 0.48 a	0.10 ± 0.00 a*	0.27 ± 0.00 b	2.16 ± 0.10 b
DF+S	13.78 ± 0.50 a	0.15 ± 0.01 a*	0.38 ± 0.01 a	19.58 ± 1.15 a
CF−S	22.16 ± 2.16 a*	0.08 ± 0.00 a	0.26 ± 0.02 b	1.80 ± 0.16 b
CF+S	18.25 ± 0.94 a*	0.07 ± 0.00 a	0.35 ± 0.01 a	28.08 ± 1.94 a*

Note: Data are presented as mean ± standard error (SE), where $n = 3$. DF−S: discontinuous flooding without additional S; DF+S: discontinuous flooding with additional S; CF−S: continuous flooding without additional S; CF+S: continuous flooding with additional S. Different letters indicate significant differences between the two S treatments under the same water management conditions and asterisks (*) indicate significant differences between different water conditions under the same S treatment ($P < 0.05$, least significant difference).

the Fe contents by 35% (−S) and 24% (+S), but decreased the Mn contents by 20% (−S) and 53% (+S) in the iron plaque on the root surface, compared with the concentrations under DF. However, neither the S nor Cd contents of the iron plaque were significantly influenced by the water management conditions. Furthermore, S addition did not have a significant effect on Fe or Mn contents but significantly increased the S and Cd contents of the iron plaque. Compared with the Cd levels without S addition, the addition of S induced 8 and 15 times the Cd content in the iron plaque under DF and CF treatments, respectively (Table 1).

2.4. Cd content in rice

Compared with the DF−S treatment, the CF+S treatment increased the biomass of rice straws and grains (Appendix A Table S1). Water conditions had a significant impact on the Cd levels in rice, and the CF regime inhibited Cd accumulation in all parts of the rice plant. The CF regime significantly reduced the Cd contents of the rice roots, straw, husks, and grains (by 88%, 88%, 91%, and 92% with S addition and by 89%, 90%, 93%, and 85% without S addition, respectively) compared with the

Cd levels under the DF regime (Fig. 3). In summary, the addition of exogenous S increased the Cd contents of all parts of the rice plant, except the grains, under CF. Under DF, the addition of S significantly increased the Cd contents (by 151%, 40%, and 72% in the rice roots, husks, and grains, respectively) compared with the contents under the −S treatment. Under CF, the addition of S increased the Cd levels by 157%, 17%, and 63% in the rice roots, straws, and husks, respectively, but decreased the Cd content (non-significantly) of the rice grains by 12% (Fig. 3).

2.5. Cd translocation and distribution in rice

Sulfur treatment significantly affected Cd transfer from the iron plaque to the rice roots and within rice, whereas water management only clearly influenced the former. The Cd translocation factor of the iron plaque to the roots under DF was approximately 9 times higher than that under CF (Table 2). Nevertheless, S addition significantly decreased the content ratio of the iron plaque to the roots (by 80% and 82% under DF and CF, respectively). In rice, S addition reduced Cd transfer from the roots to the aerial parts. The Cd translocation fac-
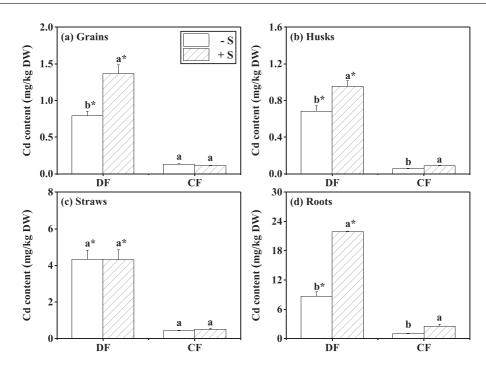
Fig. 3. – Cd contents of rice plant parts under different flooding regimes. Cd contents of (a) rice grains, (b) husks, (c) straws, and (d) roots at maturity. Data are presented as mean ± standard error (SE), where $n = 3$. DF: discontinuous flooding; CF: continuous flooding; −S: without additional S; and +S: with additional S. Different letters indicate significant differences between the two S treatments under the same water management conditions, and asterisks (*) indicate significant differences between different water management conditions under the same S treatment ($P < 0.05$, least significant difference).

Table 2. – Translocation factor of Cd in different parts of rice.

Treatment	(Root/Iron plaque)	(Aerial part/Root)	(Straw/Root)	(Husk/Straw)	(Grain/Husk)
DF−S	4.28 ± 0.23 a*	0.40 ± 0.02 a	0.50 ± 0.02 a	0.16 ± 0.01 a	1.31 ± 0.23 a
DF+S	0.84 ± 0.21 b*	0.16 ± 0.01 b	0.21 ± 0.02 b	0.22 ± 0.04 a	1.28 ± 0.03 a
CF−S	0.45 ± 0.01 a	0.38 ± 0.01 a	0.47 ± 0.02 a	0.13 ± 0.02 a	2.46 ± 0.22 a*
CF+S	0.08 ± 0.02 b	0.18 ± 0.02 b	0.22 ± 0.03 b	0.16 ± 0.01 a	1.42 ± 0.12 b

Note: Data are presented as mean ± standard error (SE), where $n = 3$. DF: discontinuous flooding; CF: continuous flooding; −S: without additional S; and +S: with additional S. Different letters indicate significant differences between the two S treatments under the same water management condition, and asterisks (*) indicate significant differences between different water management conditions under the same S treatment ($P < 0.05$, least significant difference).

tor from the rice roots to the straw significantly decreased (by more than 50%) under S addition, regardless of the water management condition (Table 2).

CF treatment effectively increased the proportion of Cd in the iron plaque. For example, compared with DF conditions, CF increased the Cd proportion in the ACA extracts by 84% (−S) and 68% (+S), but decreased it by 23% (−S) and 82% (+S) in the roots and by 29% (−S) and 74% (+S) in the shoots (Fig. 4). Additionally, compared with the −S treatments, S addition increased the proportion of Cd in the ACA extracts by 5.4 and 2.7 times and decreased it in the shoots by 43% and 80% under DF and CF, respectively. Notably, S addition had the opposite effect on the Cd proportion in rice roots under different water management conditions, slightly increasing it (23%) under DF and significantly reducing it (70%) under CF. Furthermore, S addition clearly decreased the proportion of Cd in rice

shoots. Overall, the CF and +S treatments facilitated increased Cd storage in the iron plaque (Fig. 4).

3. Discussion

3.1. Effects of water conditions and S addition on Cd dissolution

Cadmium dissolution was inhibited by flooding and promoted by drainage, which is consistent with the findings of previous studies (de Livera et al., 2011; Fulda et al., 2013; Hu et al., 2015). The experiment described herein attempted to explain the Cd dissolution mechanisms by dynamically monitoring key factors in paddy soil, including the pH, Eh, and EC values and S, Fe, and Mn concentrations (Fig. 1, Appendix A Fig. S1).
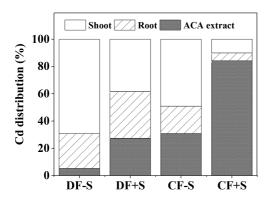
Fig. 4. – Proportion of Cd in different components of rice. Data are presented as mean ± standard error (SE), where *n* = 3. DF−S: discontinuous flooding without additional S; DF+S: discontinuous flooding with additional S; CF−S: continuous flooding without additional S; and CF+S: continuous flooding with additional S.

The Cd immobilization mechanisms after flooding can be divided into three broad categories. First, flooding causes oxygen exhaustion and mediates the reduction of OM and other substances by microorganisms (Yuan et al., 2019); thus, protons are consumed and the pH of the soil solution increases (Fig. 1c, d). Soil with a higher pH value features more adsorption sites for Cd on OM or on Fe, Al, and Mn oxyhydroxides (Sun et al., 2007), thereby increasing the Cd adsorption capacity of the soil. Previous studies have shown that a higher pH during continuous flooding leads to more negatively charged soil and increases Cd adsorption on colloids, which ultimately inhibits Cd mobility in the soil (Li and Xu, 2017). However, when the flooding recedes, oxidation processes produce protons and decrease the pH rapidly, thereby causing a large amount Cd to be released into the soil solution. Furthermore, when soil is exposed to the atmosphere, dissolved organic carbon can be converted to CO_2, which may dissolve into the soil solution as carbonic acid, leading to a further decrease in soil pH (Ponting et al., 2021). Second, this study showed that flooding clearly decreased the Eh and increased the Fe concentrations of the soil pore water (Fig. 1a, b e, f). Fe(III) is reduced to Fe(II) at a low soil Eh (de Livera et al., 2011; Yuan et al., 2019), which produces secondary iron minerals that supply abundant hydroxyl groups (Fe−OH; Li et al., 2016). The transformation of different Fe-oxide forms under flooding changes the charges of colloids and active functional groups, thereby affecting Cd adsorption on colloids (Wang et al., 2019). As indicated by a previous study, flooding can promote the conversion of crystalline iron oxides to amorphous iron oxides, which have larger surface areas and abundant reactive sites to fix Cd^{2+} (Jiang et al., 2017). Additionally, a higher pH promotes the formation of secondary Fe minerals and increases Cd fixation (Li and Xu, 2017). Finally, under long-term flooding, HS^- and S^{2-} can be generated by reducing the activity of microorganisms and forming insoluble CdS precipitates and Cd co-precipitates with Fe and Zn sulfides, which has long been considered to be important for Cd reduction under anaerobic conditions (Bingham et al., 1976). Flooding has strong inhibitory impacts on Cd dissolution, although the associated mechanisms are complex. Correspondingly, when a rice paddy is drained, the rapid oxidation of Cd-containing sulfides leads to the release of Cd^{2+}, SO_4^{2-}, Zn^{2+}, and Fe^{2+} (de Livera et al., 2011; Yuan et al., 2019). In the present study, flooding reduced the concentrations of S in soil pore water and re-draining under DF promoted the release of S, consistent with the above explanation (Fig. 1e, f).

Sulfur addition had opposite effects on Cd dissolution under the two water management conditions. Under CF, the additional S reduced Cd concentrations in the rhizosphere soil (DRC 23–158). In the middle and later stages of CF, sufficient anaerobic conditions facilitated a lower Eh and a higher pH in the paddy soil, and thus promoted the conversion of a large amount of SO_4^{2-} into S^{2-}, reducing Cd dissolution. Furthermore, the present study found that additional S significantly reduced the Fe concentrations of the rhizosphere pore water under CF (Appendix A Fig. S1e, f), demonstrating that S^{2-} primarily combines with Fe^{2+} to form FeS. Under CF, the inhibition of Fe dissolution by S addition was more obvious than the effects on Cd dissolution, indicating that Cd may have been fixed by co-precipitating with FeS. However, under DF conditions, S addition promoted Cd dissolution, especially during the drainage period (Fig. 1g, h). Owing to the increasing Eh during the drainage period of the DF treatments, sulfide would have oxidised and released a large amount SO_4^{2-} (de Livera et al., 2011). Free SO_4^{2-} can form a water-soluble complex with Cd^{2+}, leading to an increase in Cd mobility (Wang et al., 2020). Brennan and Lindsay (1996) proposed that the formation of CdS could occur when the value of pe + pH was lower than 4.85. In the present study, the pe + pH value was much higher than 4.85 during the drainage period of DF (Appendix A Fig. S1a). Moreover, the EC of the soil influences the Cd adsorption and desorption on soil colloids. In this study, the trends exhibited by S concentrations and EC in the soil pore water were similar (Fig. 1, Appendix A Fig. S1), and significant positive correlations were observed between the EC value and Cd and S concentrations (*r* > 0; *P* < 0.05). When the soil was drained, the EC value increased more rapidly under treatments with added S, indicating that SO_4^{2-} promotes ion dissociation (Appendix A Fig. S1c). It is interesting to note that, under DF, the first oxygen exposure caused SO_4^{2-} and Cd^{2+} to detach from sorption sites; however, this phenomenon was very weak during the second oxygen exposure. The Eh of the second stage still showed a significant increase, suggesting the possible presence of oxidised sulphate. Nevertheless, the pH value in the second stage of drainage was higher than that after the first drainage, which may have strengthened ion adsorption in the soil, within the more aged soil (Kayser et al., 2000). Moreover, the rice had a more developed root system and strong transpiration during the mature stage. Limited pore water under drainage conditions may be quickly taken up by the root system, which may also have influenced the sampling results. As a result, under DF, S addition increased the soil Cd concentration under the two drainage periods and the Cd content of rice grains. We speculate that the additional S may reduce Cd dissolution only under low Eh conditions caused by stable CF owing to the precipitation of CdS and coprecipitation of FeS_2 and Cd^{2+} (Feijtel et al., 1988; Wang et al., 2020). In general, the different water management conditions directly affected soil Eh and the S va-

lence state, consequentially influencing the effect of S on Cd dissolution.

The CIT method was used to synthetically identify important environmental factors affecting Cd dissolution in paddy soil. Generally, amongst the tested indicators (Eh, pH, and EC values and S, Fe, and Mn concentrations), soil Eh had the greatest effect on Cd pore water concentrations (Fig. 2a). S addition did not have clear effect when the Eh was lower than -42.5 mV, whereas it was the second most significant factor in promoting Cd dissolution in soil when the Eh was greater than -42.5 mV (Fig. 2b). These results suggest that, when Eh is low, Cd^{2+} is firmly adsorbed on colloidal substances and Fe and Mn oxides. When Eh is high, SO_4^{2-} forms and combines with Cd^{2+}, thereby reducing Cd fixation. Consequently, Eh is the primary factor that controls Cd and is critical for controlling the effect of S on Cd dissolution in soil.

In addition, the reducing effect of S on Cd concentrations was clearer in the rhizosphere soil than in the non-rhizosphere soil under CF (Fig. 1h). This may indicate that iron plaque has a binding impact on Cd (Wu et al., 2020). Moreover, the concentrations of Cd, S, Fe, and Mn in the rhizosphere soil pore water were higher than those in the bulk soil, and these deviations were more evident under DF (Fig. 1, Appendix A Fig. S1). Rice roots absorb nutrients on soil colloids or in soil pore water through transpiration, which can induce elements in the pore water to move toward the rhizosphere region (Lambers et al., 2009). Additionally, we observed rhizosphere acidification (Fig. 1c, d). Acidic organic substances secreted within the rhizosphere may activate and transfer metal ions to the rhizosphere pore water (Cao et al., 2009).

3.2. Effects of water conditions and S addition on Cd uptake and distribution in rice

In the present study, Cd accumulation in all parts of the rice plant was inhibited under CF compared to that under DF (Fig. 3). CF effectively inhibited Cd dissolution in soil (Fig. 1), which was the main mechanism through which Cd absorption and accumulation were reduced. Moreover, CF promoted the formation of iron plaque and sharply reduced the Cd transfer from the iron plaque to the rice roots (Table 1); thus, the iron plaque appeared to act as a barrier for Cd uptake and translocation to rice roots (Liu et al., 2008). In addition, flooding promoted the dissolution of Mn, which shares the Nramp5 transporter for Cd in roots (Appendix A Fig. S1). This may also have prevented the absorption of Cd by rice (Sasaki et al., 2012).

Exogenous S generally increased the Cd contents of various parts of the rice plant, especially under DF; however, the Cd content of the rice grains did not increase under CF (Fig. 3). This is consistent with the effect of S on Cd dissolution in soil. The additional S promotes Cd dissolution in soil under DF and correspondingly promotes the uptake of Cd by rice. Under CF, added S slightly reduces the Cd concentrations in the pore water and leads to increased Cd storage in the rice roots (Fig. 3). However, the role of S in plant health and Cd accumulation is complex. Our results show that the additional S significantly reduced the transfer factors of Cd from roots to straws (Table 2), decreased the proportion of Cd in the aerial parts of the rice (Fig. 4), and increased Cd storage in roots (Fig. 3). The experiment of Liu et al. (2020) found no significant effects of S

application on Cd accumulation in grain under different water regimes. The effect of S on soil chemical/biological processes influences the Cd availability for plants. However, the direct effect of S on the plant physiological responses must be also considered. The balance between the soil and plant physiological responses determines the net effect of S on Cd uptake. Different experimental conditions result in different degrees of influence of the soil and plant physiological responses, resulting in different influences between studies.

In rice plants, S is not only an essential nutrient for synthesising essential proteins but is also a structural component of several coenzymes and prosthetic groups that are necessary for normal plant growth and development (Iqbal et al., 2013). GSH and PCs are non-protein thiols that contain important functional sulfhydryl groups that play a vital role as antioxidants or chelators in plant stress tolerance and heavy metal resistance (Li et al., 2020). Additionally, if more sulfhydryl groups form complexes with Cd and are transported into vacuoles by tonoplast ATPase, Cd translocation can be limited in rice plants. Furthermore, Cd may be prevented from entering plant structural components, thereby alleviating Cd stress (Cao et al., 2018; Fan et al., 2010). S is also a constituent of the essential amino acids cysteine (Cys) and methionine (Met), which are involved in plant resistance to Cd stress (Zhang et al., 2018).

Iron plaque is an amphoteric colloid that can store Cd via adsorption and co-precipitation on the root surfaces (Lux et al., 2011). Consistent with the experimental results of Liu et al. (2008), our experiment found that the Cd content and percentage in the iron plaque both increased after flooding induced greater iron plaque formation (Table 2, Fig. 4). In contrast, our study introduced S in soil, which promoted Cd accumulation in the iron plaque, reduced Cd transfer to the aerial part, and affected the distribution of Cd. Previous studies have shown that additional S was conducive to iron plaque formation on roots and consequently affected heavy metal uptake by rice (Fan et al., 2010; Cao et al., 2018). However, this experiment did not find that additional S promoted iron plaque formation; instead, it induced Cd accumulation on the plaque and prevented Cd transfer to the roots (Table 1, Table 2). Presumably, soil environment variations affect iron plaque formation and composition; external S induced more Cd and less Fe dissolution in soil pore water, and increased soil Eh, which offset the role of S in iron plaque generation. Additionally, S changed Fe and Mn speciation; $Fe_6\text{-}XFe_x(OH)_{12}[SO_4]_{X/2}\bullet 3H_2O$ is one of the components of iron plaque (Hansel et al., 2001). Furthermore, when exogenous S was added, higher Cd proportions in the iron plaque were observed in the iron plaque–root–aerial part system (Fig. 4), further indicating the key role of iron plaque in preventing Cd from entering rice roots. Although the addition of S caused more Cd to remain in the iron plaque and hindered its transfer from the root to the aerial parts of the rice, these effects did not offset the negative influence of S on Cd dissolution in the soil under a realistic semi-aerobic environment. Under the simulation of practical water management practices (DF), additional S significantly increased the Cd content of rice grains. Therefore, regardless of the water management conditions used, the inconsiderate addition of S may increase the risk of Cd accumulation in rice grains in contaminated paddy soil.

4. Conclusions

In this study, we monitored variations in the concentrations of Fe, Mn, and S, and the soil properties (pH, Eh, or EC) under different S application and water management scenarios. Through in-depth analysis of the data, the following conclusions were drawn: (1) Eh was the primary factor affecting Cd dissolution in paddy soil. (2) In paddy soil, the effect of S on Cd dissolution depended on the soil Eh conditions. The addition of S promoted Cd dissolution when Eh was not low. (3) S application significantly improved Cd accumulation on iron plaque and prevented Cd transfer from rice roots to straw. In general, the addition of S only slightly reduced the Cd content in rice under CF, but significantly increased the Cd content under DF. The experimental results provide implications for the management of Cd-contaminated paddy soil, whereby the application of S fertiliser must be considered in light of the water management conditions to avoid increased health risks due to Cd pollution. Sulfur can influence Cd accumulation in soil and its transport in plants, and the joint action of these two processes ultimately determines the effect of S on Cd accumulation in rice grains. The effect is related to S concentration, soil properties, background element content, rice variety, agronomic management, and other factors; further in-depth research into the effects of S on Cd availability in paddy fields and the appropriate S content under different cultivation conditions is necessary to realise safe rice production.

Acknowledgments

Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2021.08.022.

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