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Organic matter addition promotes Cd immobilization in alkaline paddy soils

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Abstract

Straw incorporation into the soil is a common agricultural practice, but its effect on soil cadmium (Cd) mobility is not well understood. We added 0–20 g kg^{-1} organic matters (OMs) with different C/N ratios to three spiked alkaline paddy soils that contained a realistically low concentration of total Cd (0.94 mg kg^{-1}), and then investigated soil Cd solubility in alternate watering conditions. As current physical and chemical methods have difficulties in accurately determining the distribution and speciation of Cd in soil at a low concentration, we measured multiple soil properties to identify key factors regulating dissolved Cd concentration. For all three soils, pH and dissolved Cd concentration both decreased after flooding and increased after subsequent drying. OM addition significantly reduced soil Cd solubility at both flooding and drying stages. Random forest and linear regressions further confirmed that soil total organic carbon, rather than pH, dissolved organic carbon, or total inorganic carbon as previously suggested, was the primary predictor of Cd solubility. OMs with different C/N ratios had similar effects on soil Cd solubility, whereas the effect of OM addition rate depended on soil type. The results demonstrated the potential of straw incorporation for the remediation of Cd-contaminated alkaline paddy soils, through mechanisms that differ from those reported for acid soils.

Highlights

- OM addition promoted Cd immobilization in alkaline paddy soils after flooding and drying.
- Soil TOC (not pH, DOC, or DIC) was the primary predictor of Cd solubility.
- Effect of C/N ratio of added OM was minimal, and effect of OM addition rate depended on soil type.
- OM addition largely decreased soil pH and increased soil DOC concentration.

Keywords Alkaline paddy soil, Cadmium, C/N ratio, Flooding and drying, Organic matter

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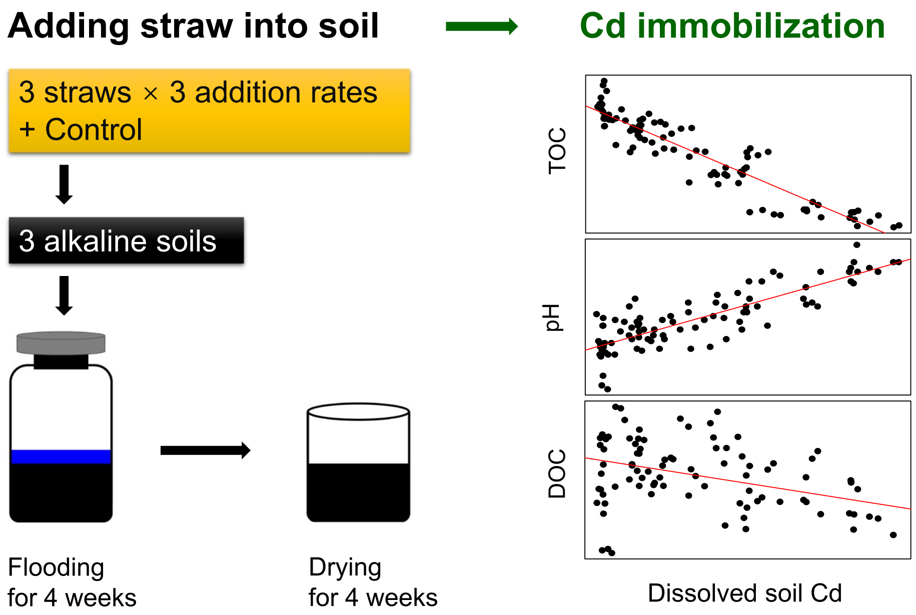
Graphical Abstract



1 Introduction

Contamination of paddy soil by cadmium (Cd) is a major issue in many countries, such as China, Korea, and Japan (Bolan et al. 2013; Yuan et al. 2021). The anthropogenic sources of Cd in paddy soils include phosphorus fertilizers, composts, mine effluents, among others (Bolan et al. 2013; Hu et al. 2016). Rice has a relatively strong ability to absorb Cd, and the excessive Cd levels in rice caused by soil contamination have raised widespread concern (Hu et al. 2016; Shi et al. 2020). In 2008, researchers found that approximately 10% of the rice samples ($n = 91$) collected from Chinese markets had a Cd concentration exceeding the Chinese Standard of 200 μg kg^{-1} for polished rice (Zhen et al. 2008), and the exceedance rate was still 10% in 2018 ($n = 160$) (Chen et al. 2018). Rice accumulates Cd mainly through root uptake (Abbas et al. 2019; Yang et al. 2020). To improve yield and facilitate harvesting, farmers usually drain the paddy field about two weeks before harvest, which is the main period for Cd uptake by rice roots (Hu et al. 2016; Huang et al. 2021). Cd accumulation by rice depends on the genetic characteristics of rice and the mobility of Cd in soil (Hu et al. 2016; Zhao and Wang 2020), and the latter is strongly influenced by soil organic matter (OM) and other soil properties (Yuan et al. 2021). The addition of OMs (e.g., crop residues and compost) into soil can therefore impact the dynamics of Cd (Park et al. 2011; Yuan et al. 2021).

Straw return is widely recommended in China because it not only returns nutrients and improves soil structure, but also avoids air pollution caused by burning and reduces carbon emissions (Li et al. 2018). However, the reported effect of straw return on soil Cd mobility is inconsistent in the literature. Straw can adsorb Cd (Yuan et al. 2019). In addition, after flooding, OM can stimulate microbial reduction processes (e.g., microbial nitrate, Fe(III), and sulfate reduction) and increase the pH of acid soils, thereby promoting the adsorption and immobilization of Cd (Yuan et al. 2019; Zhao and Wang 2020). After drainage, OM can potentially retard the oxidation of CdS and buffer soil pH change, thus possibly mitigating the release of Cd (Yuan et al. 2021). Nevertheless, it has also been reported that straw return has no significant effect on soil Cd mobility or even promotes the release of Cd (Yuan et al. 2021). The proposed reasons are that (i) organic acids released during straw decomposition lower soil pH and cause Cd desorption, and (ii) dissolved organic carbon released from straw complexes with Cd, hindering Cd adsorption by soil solid phase (Jia et al. 2010; Bai et al. 2013). The effect of OM addition on soil Cd mobility, therefore, warrants further investigation.

The remediation effectiveness of OM for Cd-contaminated paddy soils also depends on other factors, such as the C/N ratio and application rate of OM, the texture and pH of soil, etc. (Yuan et al. 2021). OM with a low C/N ratio is preferentially utilized by microorganisms because it better meets the nitrogen requirements of microorganisms (Chapin et al. 2011). Nevertheless, soil clay particles can protect OM via physical and chemical process, decreasing the availability of OM to microbes (Weil and Brady 2016; Singh et al. 2018) and presumably also the capacity of OM to adsorb Cd. Even newly added OM could be protected by clay particles (Shi and Marschner 2013). It can therefore be speculated that, if the soil has a high clay content and the OM has a high C/N ratio, the effect of OM addition on soil Cd mobility will be weaker. Unlike acid soils where the pH increases after flooding, the pH in alkaline soils decreases after flooding (Ponnamperuma 1984). Although alkaline paddy soils can also be contaminated by Cd, relevant studies are scarce (Qiao et al. 2019). Changes in Cd mobility in alkaline paddy soils during flooding and drying and the effect of OM need to be further studied in environmentally relevant conditions.

Here, OMs with different C/N ratios (rice straw, alfalfa, and the 1:1 mixture of them) were added to three Cd-spiked alkaline paddy soils with various properties. The soils contained a realistically low concentration of total Cd (but still considered contaminated) and were incubated in alternative watering conditions. Most Cd-contaminated soils have a total Cd content < 1 mg kg^{-1}, and current physical and chemical methods (e.g. synchrotron X-ray absorption spectroscopy and sequential extraction) have difficulties in accurately determining the distribution and speciation of Cd in soil at such a low concentration (Yuan et al. 2019). We therefore monitored multiple relevant soil chemical properties and explored the major factors regulating the change in dissolved Cd concentration. Our objectives were to (i) investigate how soil Cd solubility and other relevant soil properties change in different treatments after flooding and subsequent drying, and (ii) analyze the relationship between soil Cd solubility and other relevant properties to identify the major influencing factors. We tested three hypotheses: (i) OM with a low C/N ratio has a greater effect on Cd mobility than OM with a high C/N ratio, (ii) the effect of OM on Cd mobility increases with the application rate, and (iii) as soil clay particles can bind to OM, the effect of OM addition on Cd mobility will be weaker in clayey soil.

2 Materials and methods

2.1 Soils and organic matters

Three surface (0–20 cm) soils with different textures were collected from rice paddy fields in Jingzhou, Hubei Province, China, in September 2020 (Table S1). The soils were air-dried and passed through a 2 mm sieve. The air-dried soils were spiked with Cd by spraying $CdCl_2$ solution onto the soil while mixing by hand. The spiked soils were then air-dried and passed through a 2 mm sieve again (Cui et al. 2008). The spiked soil was not aged because aging has little effect on Cd availability (Smolders and Mertens 2013). The final total Cd content in the spiked soils was 0.94 ± 0.01 mg kg^{-1}. The soil Cd content of ~ 1 mg kg^{-1} exceeds the risk screening values for Cd in paddy soil, which are 0.3 mg kg^{-1} when soil pH ≤ 5.5, 0.4 mg kg^{-1} when $5.5 <$ soil pH ≤ 6.5, 0.6 mg kg^{-1} when $6.5 <$ soil pH ≤ 7.5, and 0.8 mg kg^{-1} when soil pH > 7.5, given in the Chinese standard GB 15618—2018. And for ~ 1 mg Cd kg^{-1} soil, Cd content in rice may exceed 0.2 mg kg^{-1} (the Chinese limit for Cd concentration in rice) (Zhao and Wang 2020). The OMs used in this study were dried rice straw and alfalfa (Table S2), which were ground and passed through a 1.5 mm sieve. The properties of soils and OMs were determined before being used in the experiment (Tables S1 and S2).

2.2 Experimental setup

Ten treatments (each with three replicates) were included for each of the three soils: control (without OM addition) and the addition of three OMs with different C/N ratios (rice straw, alfalfa, and a 1:1 mixture of rice straw and alfalfa with C/N ratios of 50, 11, and 30, respectively) each at a rate of 0.5%, 1%, or 2% (w/w, oven-dry basis). So, there were 30 treatments in total. After thoroughly mixing the soil with the OM in a plastic bag (Yuan et al. 2019), 10 g of soil was placed into a 60 mL serum bottle for each treatment, and then 10 mL Milli-Q water was added such that after saturation there was approximately 5 mm overlying water. The oxygen in the headspace of bottles was displaced by blowing nitrogen for 10 min. The bottles were then quickly sealed with rubber stoppers and aluminum lids and incubated at 28 °C in darkness. After 4 weeks of anaerobic incubation, the soil in the bottles was transferred to 50 mL polypropylene cups and rapidly dried in a fan-forced oven at 30 °C to reduce the water content to 50% of soil water holding capacity (WHC) to initiate aerobic incubation. [A water content of 40–60% of the WHC is optimum for most aerobic processes (Wilke 2005). The cups, loosely covered with lids, were then incubated at 28 °C in darkness for another 4 weeks. The soil water content was maintained by adding drops of Milli-Q water every 2–3 days. Soil WHC was determined according to Wilke (2005).

2.3 Sampling and chemical analyses

Three soil microcosms for each treatment were sacrificed in weeks 0, 4, and 8, and soil sampling and chemical analyses were conducted mainly according to Yuan et al. (2019). Briefly, after 4 weeks of anaerobic incubation, the bottles were transferred into a glove box. The soil paste in each bottle was decanted into a 50 mL polypropylene cup and stirred thoroughly. For analyses of dissolved Cd, Fe, C, and N, approximately 1 g of the soil paste was weighed into a 50 mL centrifuge tube, and 20 mL of 0.01 M $CaCl_2$ solution was added inside the glove box (Houba et al. 2000). Then, the tubes were closed tightly and shaken end-over-end at 50 rpm and room temperature for 1 h outside the glove box. After centrifugation at 7800 rpm for 2 min, the tubes were then transferred back into the glove box, and the supernatants were passed through 0.22 μm membrane filters. The extracts were acidified by adding 0.2 mL of 1 M HCl to 20 mL of the filtered supernatant in the glove box (Houba et al. 2000). The chemical solutions employed in the anoxic extraction were deoxygenated before being applied to bubbling ultrapure N_2 overnight. Dissolved Cd and Fe^{2+} concentrations in the acidified extracts were then determined with an inductively coupled plasma-mass spectrometry (ICP-MS) system (7900, Agilent, USA) and the o-phenanthroline photometry method (Fadrus and Maly 1975). Dissolved organic carbon (DOC) and dissolved total nitrogen (DTN) were determined with a TOC analyzer (Multi N/C 3100, Analytic Jena, Germany). No interference between Ca^{2+} and DOC was observed when determining the concentration of soil DOC with 0.01 M $CaCl_2$ as an extraction reagent (Table S3 and Fig. S1). Ammonium-nitrogen (NH_4^+-N) and nitrate-nitrogen (NO_3^--N) were measured using a continuous flow analyzer (AA3, SEAL, Germany). Dissolved organic nitrogen (DON) was calculated by subtracting NH_4^+-N and NO_3^--N from DTN. For measuring soil pH, Eh, and water content, approximately 3 g of the soil paste was transferred into a 15 mL centrifuge tube. Soil pH and Eh were recorded after inserting pH and Eh electrodes (Model IJ44A and IJ64D, Ionode, Australia) into the soil paste outside the glove box, and the remaining soil paste was then used to determine soil water content by drying at 105 °C for at least 48 h. Soil pH was, therefore, measured at a soil to water ratio of 1:1. Another portion of soil paste was freeze-dried and passed through a 0.149 mm sieve. Then, soil total organic carbon (TOC) and total inorganic carbon (TIC) were determined using the Walkley–Black method (Nelson and Sommers 1996) and the modified pressure-calcimeter method (Sherrod et al. 2002), respectively.

In week 0 and week 8 (4 weeks after aerobic incubation), sampling and chemical analyses were performed outside the glove box. For the pH and Eh measurement, Milli-Q water was added to the soil samples to produce soil paste that had the same water content as the soil samples in week 4.

2.4 Statistical analyses

Two-way analysis of variance (ANOVA) and post hoc pairwise comparisons with Bonferroni adjustment were conducted to analyze the effects of factors on the concentration of 0.01 M $CaCl_2$ extractable Cd (hereinafter referred to as dissolved Cd). When time was included in ANOVA, time was considered as a repeated measures factor. Because of the small sample size (number of replicates = 3), three-way ANOVA (soil, OM addition, and time as a repeated measures factor) was not adopted (Cohen 2013). Pearson correlation and linear regression analyses were conducted to explore the relationships of dissolved Cd concentration with other soil properties. Random forest regression was performed according to Jiao et al. (2018) to identify significant predictors of soil Cd solubility. ANOVA was conducted with IBM SPSS Statistics 23 (IBM Co., Armonk, NY, USA), and other statistical analyses were performed in R (version 4.1.1).

3 Results and discussion

3.1 Change in soil Cd solubility and major influencing factors

In week 0 (before flooding), for each soil, the concentrations of dissolved Cd in the ten treatments were basically the same; among soils, dissolved Cd concentration was the highest in the sandy loam (average of 12.85 $\mu g\ kg^{-1}$), followed by the silt loam (average of 8.19 $\mu g\ kg^{-1}$) and the silty clay (average of 2.37 $\mu g\ kg^{-1}$) (Fig. 1). After flooding, soil Eh decreased from approximately 180 mV in week 0 to approximately -180 mV in week 4 (soil Eh in the unamended sandy loam was expected to be 40 mV in week 4) (Fig. S2), and pH decreased from 7.4–8.0 in week 0 to 6.6–7.8 in week 4 across all 30 treatments (Fig. S3). Meanwhile, dissolved Cd concentration decreased to 1.67–4.73 $\mu g\ kg^{-1}$ (by 26%-60% compared to week 0) in the unamended control soils, whereas to below the detection limit when OM was added (regardless of addition rate or C/N ratio) (Fig. 1). After drying, across all 30 treatments, soil Eh increased to approximately 130 mV (Fig. S2), pH rose to 7.6–8.2 (Fig. S3), and dissolved Cd concentration increased to 0.70–9.63 $\mu g\ kg^{-1}$ in week 8 (Fig. 1). When > 0.5% OM was added into the soils, dissolved Cd concentration in week 8 was lowered by 23%-73% compared to the controls. In most cases, the concentration of dissolved soil Cd decreased with the increasing addition rate of OM, whereas it did not significantly differ when OMs with different C/N ratios were added (Fig. 1). The results demonstrated that OM addition effectively promoted the immobilization of Cd after flooding and subsequent drying.

To explore the mechanisms of change in soil Cd mobility, we analyzed the relationships between dissolved Cd

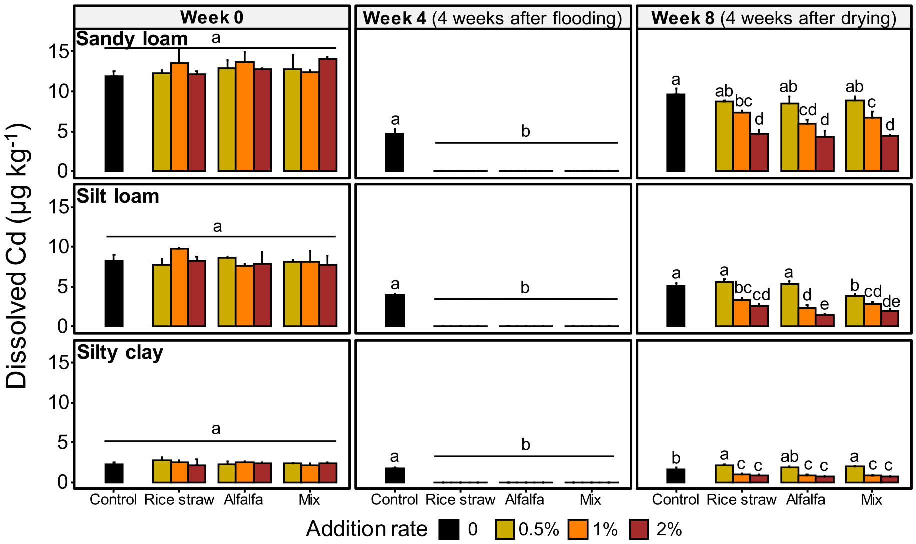


Fig. 1 Dissolved Cd concentration in three alkaline paddy soils with different textures after 4 weeks of flooding and subsequent 4 weeks of drying. Mix was a 1:1 mixture of rice straw and alfalfa. In week 4, dissolved Cd concentration was below the detection limit in all treatments with OM addition. For each soil and time point, values that do not share the same letter are significantly different ($P < 0.05$)

concentration and other soil chemical properties. After 4 weeks of flooding, because the dissolved Cd concentration in all treatments with OM addition decreased to under the detection limit, only the dissolved Cd concentration in the unamended control soils was analyzed, and it had a significant negative correlation with soil TOC ($R^2 = 0.674$, $P < 0.01$, $n = 9$) (Table S4). The data after 4 weeks of drying ($n = 90$) were analyzed with random forest regression; again, TOC was the most important factor influencing dissolved Cd concentration, followed by pH (Fig. 2a). Correlation tests also showed that dissolved Cd concentration had a negative relationship with TOC ($R^2 = 0.869$, $P < 0.01$, $n = 90$) and a positive relationship with pH ($R^2 = 0.699$, $P < 0.01$, $n = 90$) (Fig. 2b-c and Table S5). We further found that TOC had a better linear fit than pH to the concentration of dissolved soil Cd (Table 1).

Soil TOC is the primary factor regulating soil Cd mobility in this study (Fig. 2 and Table 1), which seems contrast with the general belief that soil pH is the primary factor controlling dissolved Cd concentration. Sauvé et al. (2000) collected data from over seventy papers and constructed empirical models to predict dissolved Cd concentration in the soil; they found that pH played a major role in predicting dissolved Cd concentration, whereas the contribution of TOC was minor.

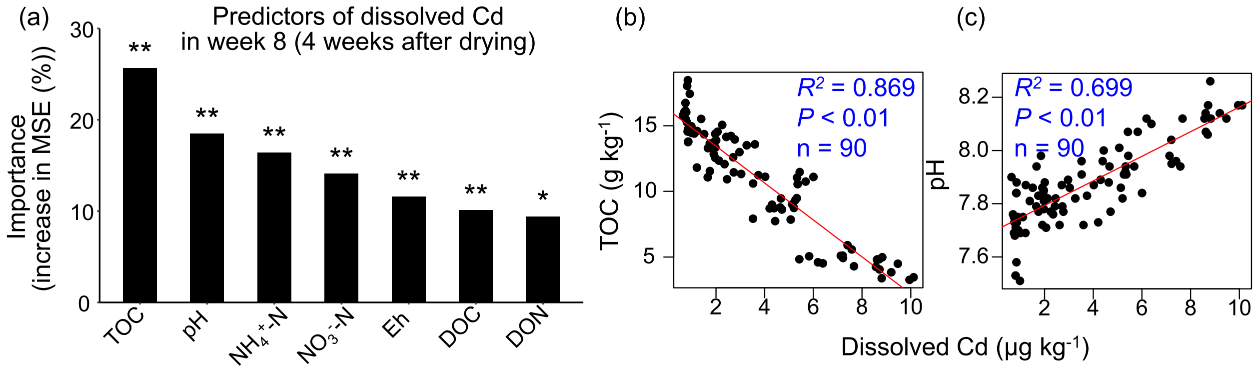


Fig. 2 **a** Significant predictors of dissolved Cd concentration after 4 weeks of drying identified by random forest regression ($n = 90$). Data for the predictors are given in Figs. S2-S8 and Table S8. [Dissolved Fe^{2+} concentration was not included because it was below the detection limit in all treatments in week 8 (Fig. S9)]. MSE indicates mean squared error. *, $P < 0.05$; **, $P < 0.01$. **b**, **c** Correlations of soil TOC concentration and $pH_{(1:1\ soil:water)}$ with dissolved Cd concentration after 4 weeks of drying

Table 1 Coefficients and R^2 for the linear regressions of dissolved Cd concentration (μg kg^{-1}) against soil $pH_{(1:1\ soil:water)}$, TOC (g kg^{-1}), or both in week 8 (4 weeks after drying) ($n = 90$)

Dependent variable	Intercept	Coefficient		R^2
$\log_{10}$ (dissolved Cd) =	1.937 ± 0.095		- 1.491 ± 0.094 × $\log_{10}$ (TOC)	0.743***
$\log_{10}$ (dissolved Cd) =	- 14.018 ± 1.224	+ 1.837 ± 0.155 × pH		0.614***
$\log_{10}$ (dissolved Cd) =	- 1.123 ± 2.132	+ 0.359 ± 0.250 × pH	- 1.262 ± 0.185 × $\log_{10}$ (TOC)	0.749***

***, $P < 0.001$

However, soil pH varied from 3 to 9, and most of the data were from acidic soils as stated by previous study; this differs from our study, where pH ranged from 6.6 to 8.2. It is well known that OM is an efficient adsorbent of Cd (Smolders and Mertens 2013), and adding OM can improve the adsorption capacity of soil for Cd (Yuan et al. 2019). Indeed, after shaking 0.4 g of soil from each treatment with 25 mL of $CdCl_2$ solution containing 1000 μg L^{-1} Cd^{2+} for 24 h, we found that the dissolved Cd concentration in the treatments with OM addition was 3%-20% lower compared to that in the controls, and was lower when more OM was added (Table S6). Furthermore, the specific surface area and functional groups of straw may undergo changes, and its ability to adsorb cations may increase after decomposition (Li et al. 2014; Li and Zhong 2021).

Soil pH is a secondary factor affecting Cd mobility in this study. In week 0, soil pH was lower than that in the controls when OM was added (Fig. S3), probably because the pH of rice straw and alfalfa was lower than that of the soils (Tables S1 and S2). In weeks 4 and 8, in the treatments with OM addition, soil pH remained lower than or was not significantly different from that in the controls (Fig. S3). In contrast to acid soils, the pH of alkaline soils decreases after flooding due to the accumulation of CO_2 and increases after drying (Fig. S3) (Ponnamperuma 1984). In acid soils, the increase in pH typically promotes the immobilization of Cd because H^+ can effectively compete with Cd^{2+} for adsorption sites (Smolders and Mertens 2013). In the range of soil pH in this study (6.6–8.2), pH increase, however, was accompanied by Cd mobilization (Figs. 1 and S3). Zhao et al. (2020) also observed this trend in an alkaline paddy soil when studying the effect of silicon on Cd uptake by rice. Regarding the change in soil Cd mobility with time, the dissolved Cd concentration in all treatments was lower in week 4 than week 0 (Fig. 1), possibly because the decrease in soil pH after flooding had a relatively small impact on Cd mobility compared to other processes such as Cd adsorption by OM. As the pH scale is logarithmic, a unit change in pH in our soils, compared to acid soils, means a much smaller change in the concentration of H^+. Dissolved Cd concentration was higher in week 8 than week 4 (Figs. 1 and S3). The contribution of OH^- to this increase in Cd solubility was expected to be limited because OH^- does not play a major role in Cd complexation when $pH < 9$ (Ford et al. 2007). We speculate that the increase in Cd solubility was because of the formation of dissolved Cd complexes with inorganic ligands such as CO_3^{2-}, Cl^-, and SO_4^{2-} (Christensen and Haung 1999; Ford et al. 2007). These inorganic ligands may be released during the decomposition of added OM after soil drying, but this cannot be confirmed by the data obtained and requires future study. The reason for the positive relationship between dissolved Cd concentration and soil pH in week 8 (Fig. 2c) may be that OM adsorbed Cd and at the same time decreased soil pH (Figs. 1 and S3).

Some low-molecular-weight organic acids can also complex Cd^{2+} and promote the desorption of Cd (Christensen and Haung 1999). This has been taken by some researchers to explain the increased Cd availability in soil and Cd uptake by plants after OM addition (Yuan et al. 2021). In this study, OM addition significantly increased soil DOC concentration compared to the controls in week 0 (Fig. S5), because the added OM released DOC (Table S2). However, in weeks 4 and 8, the effect of OM addition on soil DOC concentration was small (Fig. S5), and there was no positive correlation between DOC and dissolved Cd concentration after flooding or drying (Tables S4 and S5). Many studies also showed that DOC concentration diminished in OM-amended soil after a period of incubation (Ye and Horwath 2017; Gao et al. 2018; Zuo et al. 2022), and a minimal effect of soil DOC on dissolved Cd concentration was also observed by Cui et al. (2008).

Khaokaew et al. (2011) reported that Cd was immobilized in carbonates during flooding in an alkaline paddy soil with 142 mg kg^{-1} Cd. Our soils contained only 0.94 mg kg^{-1} Cd, so it was most unlikely that $CdCO_3$ was formed, considering a solubility product of 10^{-12} for $CdCO_3$ (Smolders and Mertens 2013). Indeed, there was no negative correlation between soil TIC and dissolved Cd concentration after flooding or drying (Tables S4 and S7).

Compared to the controls, OM addition significantly promoted iron reduction after flooding, generating a substantial concentration of dissolved Fe^{2+} (Fig. S9). It was reported that the secondary iron minerals formed during iron reduction could immobilize Cd by adsorption and co-precipitation (Muehe et al. 2013a, 2013b; Li et al. 2016). Since a realistically low concentration of total soil Cd (~ 1 mg kg^{-1}) was employed in this study, we could not accurately characterize the distribution of Cd in the soil solid phase (Yuan et al. 2019). Nevertheless, the contribution of secondary iron minerals to Cd immobilization might be limited in this study. Compared to the controls, adding rice straw to soil at 0.5% hardly affected Fe^{2+} concentration but significantly reduced dissolved Cd concentration in week 4 (Figs. 1 and S9).

The precipitation of CdS can also decrease soil Cd solubility (Zhao and Wang 2020), but CdS precipitation in our soils, which had low Cd and S content (Table S1), probably was limited. Considerable CdS precipitation occurs only when the soil has high concentrations of Cd and reducible sulfate (e.g., ~ 20 mg kg^{-1} Cd and > 2 mmol kg^{-1} sulfate) and low concentrations of other chalcophile metals (e.g., Cu), which also precipitate with sulfide (Fulda et al. 2013).

3.2 Effects of C/N ratio and application rate of OM as well as soil texture

Soils amended with alfalfa, which had a lower C/N ratio, showed greater OM decomposition (TOC decrease) than soils amended with rice straw, which had a higher C/N ratio, especially at the 0.5% and 1% OM addition rates (Fig. S4). TOC in soils amended with alfalfa and rice straw decreased on average by 2.89 and 1.90 g kg^{-1} between weeks 0 and 8, across soils and OM addition rates. Nevertheless, in week 4, in all treatments with OM addition, regardless of the C/N ratio of OM, the concentration of dissolved Cd was under the detection limit. In week 8, the concentration of dissolved Cd in treatments with alfalfa was marginally lower than that in treatments with rice straw (on average by 13% across soils and OM addition rates) (Fig. 1). Therefore, we can largely reject the hypothesis (i): OM with a low C/N ratio has a greater effect on Cd mobility than OM with a high C/N ratio. This suggests that for Cd immobilization in paddy soil, addition of rice straw is effective, and addition of OM with a lower C/N ratio is not needed.

Within a certain range, increasing the amount of added OM may better facilitate soil remediation (Yuan et al. 2015; Muhammad et al. 2021). Khan et al. (2018) found that higher OM application rates caused greater decrease in available Cd concentration in mine-impacted soils, consistent with our results in the sandy loam and silt loam in week 8 (Fig. 1). However, it was not always better to add more OM. In the silty clay, dissolved Cd concentrations in the treatments with 1% and 2% OM were basically the same (Fig. 1). This inconsistency indicates that the effect of OM addition rate depends on soil type. Based on the above results, we can partially accept the hypothesis (ii): the effect of OM on Cd mobility increases with the application rate.

We calculated the absolute decrease (in μg kg^{-1}) in dissolved Cd concentration in treatments with OM addition compared to the controls in week 8, which was smaller in a soil with a higher clay content (see also Fig. 1). The adsorption experiment also showed that after adding the same type and amount of OM to the soil, the increase in Cd adsorption was smaller in a soil with a higher clay content (Table S6), probably because clay particles bound to OM and decreased its capacity to adsorb Cd. Therefore, we can accept the hypothesis (iii): the effect of OM on Cd mobility will be weaker in clayey soil. Nevertheless, no clear relationship was found between soil texture and OM decomposition in this study (Fig. S4). It should also be noted that the initial dissolved Cd concentration in week 0 was already lower in a soil with a higher clay content. A soil with a higher clay content also had a higher TOC content (Fig. S4), and both OM and clay particles can adsorb Cd. Thus, the above results do not suggest that with the same pollution load, more OM should be added to a clayey soil than a sandy soil for Cd immobilization, because after the same amount of Cd enters the soil, the dissolved Cd concentration in a clayey soil will be lower than that in a sandy soil due to Cd adsorption by clay particles (see also Table S6 and data in week 0 in Fig. 1). Rather, our results suggest that to achieve the same decrease in soil Cd solubility, more OM should be added to a soil with a high clay content.

4 Conclusions

Our results showed that in both anaerobic and aerobic conditions, OM addition effectively reduced Cd solubility in various alkaline paddy soils containing a realistically low concentration of total Cd (~ 1 mg kg^{-1}), through mechanisms that differed from those reported in acid soils. This study, along with our previous investigation in an acid paddy soil (Yuan et al. 2019), demonstrates the potential of straw return for the remediation of Cd-contaminated paddy soils. These results were obtained in soil systems without plants. In paddy fields, however, straw return can sometimes cause Cd mobilization (Yuan et al. 2021). Future studies should explore the role of plants in Cd mobilization, because if there is no plant, straw return will not cause Cd mobilization in soil.

Abbreviations

DOC	Dissolved organic carbon
DON	Dissolved organic nitrogen
DTN	Dissolved total nitrogen
OM	Organic matter
TIC	Total inorganic carbon
TOC	Total organic carbon

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1007/s44246-023-00072-2.

Additional file 1: Table S1. Information for paddy soils used. **Table S2.** Selected properties of plant residues used. **Table S3.** Soil DOC (dissolved organic carbon) extracted by H_2O and 0.01 M $CaCl_2$ from 9 soil treatments. There was no significant difference between DOC concentrations obtained by the two extraction reagents. **Table S4.** Correlation matrix of soil factors in week *4* (after 4 weeks of flooding). (Pearson; $n = 9$ (only data for the unamended control soils were shown because in all soils with added OM the dissolved Cd concentration was under detection limit); only R values with adjusted $P < 0.05$ were shown). **Table S5.** Correlation matrix of soil factors in week *8* (after 4 weeks of drying). (Pearson; $n = 90$; only R values with adjusted $P < 0.05$ were shown). **Table S6.** Effects of OM addition on Cd adsorption by soil. Data are dissolved Cd concentrations (μg L^{-1}) after 0.4 g of soil from each treatment was shaken with 25 mL of $CdCl_2$ solution containing 1000 μg L^{-1} Cd^{2+} for 24 h. Control, no OM added; Mix, 1:1 mixture of rice straw and alfalfa. **Table S7.** Correlation matrix of soil factors (*including TIC*) in week *8* (after 4 weeks of drying). (Pearson; $n = 77$; only R values for adjusted $P < 0.05$ were shown). **Table S8.** Soil properties measured in this study. **Fig. S1.** Correlation between concentrations of soil DOC extracted by H_2O and 0.01 M $CaCl_2$. For details, see Table S3. **Fig. S2.** Eh in three alkaline paddy soils with different textures after 4 weeks of flooding and subsequent 4 weeks of drying. Mix was a 1:1 mixture of rice straw and alfalfa. For each soil and time point, values that do not share the same letter are significantly different ($P < 0.05$). **Fig. S3.** $pH_{(1:1\ soil:water)}$ in three alkaline paddy soils with different textures after 4 weeks of flooding and subsequent 4 weeks of drying. Mix was a 1:1 mixture of rice straw and alfalfa. For each soil and time point, values that do not share the same letter are significantly different ($P < 0.05$). **Fig. S4.** TOC (total organic carbon) concentration in three alkaline paddy soils with different textures after 4 weeks of flooding and subsequent 4 weeks of drying. Mix was a 1:1 mixture of rice straw and alfalfa. For each soil and time point, values that do not share the same letter are significantly different ($P < 0.05$). The soil TOC for week 0 was calculated based on the soil TOC and carbon content of added organic matter, so no multiple comparisons were conducted. **Fig. S5.** DOC (dissolved organic carbon) concentration in three alkaline paddy soils with different textures after 4 weeks of flooding and subsequent 4 weeks of drying. Mix was a 1:1 mixture of rice straw and alfalfa. For each soil and time point, values that do not share the same letter are significantly different ($P < 0.05$). **Fig. S6.** DON (dissolved organic nitrogen) concentration in three alkaline paddy soils with different textures after 4 weeks of flooding and subsequent 4 weeks of drying. Mix was a 1:1 mixture of rice straw and alfalfa. For each soil and time point, values that do not share the same letter are significantly different ($P < 0.05$). **Fig. S7.** NO_3^--N concentration in three alkaline paddy soils with different textures after 4 weeks of flooding and subsequent 4 weeks of drying. Mix was a 1:1 mixture of rice straw and alfalfa. For each soil and time point, values that do not share the same letter are significantly different ($P < 0.05$). **Fig. S8.** NH_4^+-N concentration in three alkaline paddy soils with different textures after 4 weeks of flooding and subsequent 4 weeks of drying. Mix was a 1:1 mixture of rice straw and alfalfa. For each soil and time point, values that do not share the same letter are significantly different ($P < 0.05$). **Fig. S9.** Dissolved Fe^{2+} concentration in three alkaline paddy soils with different textures after 4 weeks of flooding and subsequent 4 weeks of drying. Mix was a 1:1 mixture of rice straw and alfalfa. In weeks 0 and 8, dissolved Fe^{2+} concentration was below the detection limit in all treatments. In week 4, for each soil, values that do not share the same letter are significantly different ($P < 0.05$). **Fig. S10.** Significant soil predictors (*including TIC*) of dissolved soil Cd in week 8 (after 4 weeks of drying) identified by random forest regression. In week 4, because the concentration of dissolved Cd in all soils with added OM was below the detection limit, random forest regression was not conducted. Dissolved Fe^{2+} concentration was not included because it was below the detection limit in all treatments in week 8. For 13 out of the 90 samples in week 8, there was not enough soil left for TIC (total inorganic carbon) determination, sample numbers were 77. MSE indicates mean squared error. Significance levels: *, $P < 0.05$; **, $P < 0.01$. For abbreviations of soil predictors, see Table S4.

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Authors' contributions
All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Zhaoyang Sun, Wenjun Zhang, Jiaping Wang, and Qiusheng Chen. The first draft of the manuscript was written by Chaolei Yuan and Zhaoyang Sun and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials
Data will be available from the corresponding author on reasonable request.

Declarations

Competing interests
Hongwen Sun is an editorial board member of Carbon Research and was not involved in the editorial review of, or the decision to publish, this article. All authors declare that there are no competing interests.

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