



# Effect of Organic Amendments on Cadmium Bioavailability in Soil and its Accumulation in Rice Grain

Khandoker Qudrata Kibria<sup>1</sup> · Md. Azharul Islam<sup>2</sup> · Sirajul Hoque<sup>3</sup> · Mohammad Zaber Hossain<sup>1</sup> · Md. Atikul Islam<sup>4</sup>

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## Abstract

A pot trial was conducted during the boro (dry) season to evaluate the impact of six traditional organic amendments (OAs) on the growth of SL-8 rice variety in both agricultural and cadmium (Cd) stressed soil at 2% and 4% application rates. Traditional OAs used in the study were cow dung, mustard oil cake (MOC), rice husk, saw dust, tea leaf and vermi compost (VC). Except for cow dung all other OAs were found to remove 99% of Cd from the aqueous solution, while cow dung removed 95%. Rice grain grown in OA-added soil in all application rates contained less Cd than the control. A 2% application rate was found to be more effective in reducing both Cd bioavailability and Cd in grain. OA application in soil significantly influenced soil pH in all cases. Though both bioavailable Cd in soil and grain Cd were reduced by the OA addition, the Cd uptake tendency of SL-8 rice variety markedly increased because of Cd spiking in soil.

**Keywords** Grain Cd · Organic amendments · Rice grain · Soil bioavailable Cd · Soil pH

## Introduction

Cadmium (Cd) is a toxic heavy metal, commonly bioavailable as Cd<sup>2+</sup> in soil and is non-essential for crops. Cd is added to soil through both natural and anthropogenic

processes (Reboredo et al. 2019; Gil et al. 2022). Uptake of Cd from soil solution is the point of entry of this element into the food chain (Moreno-Jimenez et al. 2016; Wei et al. 2019). Suzuki et al. (1988) claimed that Cd-rich food is usually produced in Cd-rich soil. Food from plant sources such as grain and vegetable are the major sources of Cd for humans (WHO 2015; Song et al. 2017). Bangladesh is a country where significant Cd addition to crop land is practiced (Bhattacharyya et al. 2008; Islam et al. 2018). Once incorporated into the soil it is difficult to withdraw the metal and its further degradation occurs (Jiang et al. 2012). A Cd concentration between 0.83 and 4.08 mg/kg (mean 2.17 mg/kg) has been detected in agricultural surface soils of Bangladesh (Mamun et al. 2021). Rice grain accumulates Cd and then transfers it to consumers (Li et al. 2017). This is a major human food safety issue worldwide and especially in countries like Bangladesh where per capita rice consumption is the highest, i.e. 181.3 kg/year (FAO 2020).

Of the various dietary Cd sources rice consumption is the major one for the people of Bangladesh. Long-term rice consumption that contains more than 0.2 mg/kg Cd may irreversibly cause kidney damage, cartilage disease, liver dysfunction, etc. (Zhu et al. 2020). Cd deposition in rice grain is shaped by many factors like rice varieties (Kibria et al. 2022), Cd bioavailability (Li et al. 2017), soil pH (Hou et

✉ Md. Atikul Islam  
atik@es.ku.ac.bd; atikku\_es@yahoo.com

Khandoker Qudrata Kibria  
kibriaku@gmail.com

Md. Azharul Islam  
iazharul@gmail.com

Sirajul Hoque  
sirajswed@du.ac.bd

Mohammad Zaber Hossain  
zaberhossain74@yahoo.com

<sup>1</sup> Soil, Water and Environment Discipline, Khulna University, Khulna 9208, Bangladesh

<sup>2</sup> Forestry and Wood Technology Discipline, Khulna University, Khulna, Bangladesh

<sup>3</sup> Department of Soil, Water and Environment, University of Dhaka, Dhaka 1000, Bangladesh

<sup>4</sup> Environmental Science Discipline, Khulna University, Khulna, Bangladesh

al. 2021), organic matter (OM) in soil (Filipovic et al. 2018; Zhu et al. 2020), redox potential, microbial activity (Zhu et al. 2020), etc. Hence, new strategies need to be devised urgently to minimize bioavailable Cd in soil so that it does not appear in rice grain.

The application of organic amendments (OA) to reduce solubility and bioavailability of toxic metals has increased worldwide (Song et al. 2021). OA provides plants with essential nutrients, while improving soil's physical, chemical and biological properties (Saengwilai et al. 2020). OA added to soil immobilize metallic ions primarily by adsorption, ion exchange, surface complexation and precipitation (Hamid et al. 2019). Colloidal properties of OA especially ion exchange capacity and effective surface area enhance adsorption of metals on their surfaces and subsequently reduce metal bioavailability (Hamid et al. 2018). Of all soil properties soil pH noticeably influences Cd bioavailability (Sauve et al. 2000). Organic wastes like farmyard manure, compost, VC, biochar, crop straw and sewage sludge are commonly applied as OA in soil to provide plants with nutrient and immobilize Cd (Liu et al. 2020). Several authors confirm the incorporation of crop straw in soil to affect metal bioavailability to plants (Wang et al. 2015; Yin et al. 2016). Mohamed et al. (2010) stated that the use of rice straw in soil elevates soil pH and ultimately minimizes Cd bioavailability by binding the water soluble bivalent Cd<sup>2+</sup> ion. Bioavailable Cd reduction was also reported by using corn and rape straw in an eight year-long field experiment by Rui et al. (2020). Other researchers used cattle manure (Wang et al. 2012), farmyard manure (Grüter et al. 2017), compost (Pardo et al. 2014), animal waste (Sato et al. 2010), etc., as OA to reduce bioavailable Cd in agricultural and polluted soil. VC was also found as an effective agent to curtail bioavailable Cd by both adsorption and forming organo-metallic complexes (Zhang et al. 2019). The effectiveness of

OA to diminish Cd bioavailability depends on many factors such as nature of the OM (Li et al. 2018), stages of decomposition (Putwattana et al. 2015), application rate, application duration (Rui et al. 2020), soil type (Shi et al. 2011), soil pH (Zhu et al. 2020; Jiale et al. 2021), etc. Hamid et al. (2019) reported the availability of amendments as one of the key factors that can minimize Cd bioavailability in soil.

No effective measures have been operated in the soils of Bangladesh so far to minimize Cd availability for plants' uptake. There is insufficient information on the effectiveness of traditional OA to reduce Cd availability of rice grain grown in the soils of Bangladesh. Moreover, information about the best rate of OA application for agriculture in Cd contaminated soil is not available (Mamun et al. 2021). Meharg et al. (2013) studied rice grain Cd of 12 countries and found that the maximum Cd (99 µg/kg) existed in grains from Bangladesh. The present study is designed to observe the influence of widely used traditional OA on Cd bioavailability in both agricultural and Cd-stressed soil, and its effect on grain Cd concentration in rice. The effect of OA application rate (2% and 4%) was also measured in this study. Most soils in Bangladesh have less than 1.5% and some soils have even less than 1% OM (FRG 2018). The recommended doses (not more than 0.25%) of organic fertilizer for agricultural land is very low (FRG 2018). Consequently, the outcome of the study will provide information on the effectiveness of traditional OA and the best application rate required to reduce Cd in rice grain.

## Materials and Methods

Surface soil (0–20 cm depth) was collected from multiple sites to prepare a composite sample from a rice-growing field (22.802° N, 89.533° E). The land has served to cultivate rice for the last 20 years. After air drying the soil was crushed using a wooden hammer so that the sample could pass through a 4 mm sieve for use in pots. For laboratory determination soil was ready to pass through a 2 mm sieve. Properties of the soil are summarized in Table 1. The soil is non-saline and neutral in pH. It was clay textured and the clay content amounted to 58.74%. Lower CEC (45 cmol/kg) means that the clay fraction is composed mostly of less active clay. Cd content of the soil was a bit higher (6.16 mg Cd/kg soil) probably due to the application of agrochemicals for 20 consecutive years during rice cultivation. Cd distribution data reveal that most Cd is present in an OM bound fraction followed by MnO bound > FeO bound > Exchangeable > Carbonate bound. A small amount of Cd (2.44% of total) was found in the bioavailable pool.

Six OA, namely cow dung, MOC, rice husk, saw dust, tea leaf (residue left after tea preparation) and VC were used

**Table 1** Basic soil properties of soil used in the experiment

Soil parameters	Values
Soil textural class	Clay
Sand	14.31%
Silt	26.95%
Clay	58.74%
pH	7.23
EC	1.10 mS
Organic matter	2.73%
Cation exchange capacity	45 cmol/kg
Total Cd	6.16 mg/kg
Bioavailable Cd	0.15 mg/kg
<b>Cd fractions</b>	
Exchangeable Cd	0.09 mg/kg
Carbonate bound Cd	0.06 mg/kg
MnO bound Cd	0.62 mg/kg
FeO bound Cd	0.41 mg/kg
Organic matter bound Cd	1.79 mg/kg

in the experiment. Collected OAs were air dried in a shed, ground and passed through a 2 mm sieve. Some basic data of the OAs are given in Table 2. Total Cd content of cow dung is high (2.35 mg/kg) whereas the other OAs contain low Cd (0.05 mg/kg). Bioavailable Cd content of the OAs is very low (0.001 mg/kg) and this indicates that most of the Cd exists in organic combination and not subject to exchange. CEC of the materials varied considerably (68–310 cmol/kg).

Six OA used in this study to reduce Cd bioavailability in soil was first tested in a laboratory to assess their Cd<sup>2+</sup> removal efficiency from aqueous solution. Standard Cd solution (1000 mg/L) was prepared by dissolving CdCl<sub>2</sub>.H<sub>2</sub>O (Sigma-Aldrich, Germany). As soil solution contains less Cd, for example 5 µg/L (Kubier et al. 2019) by diluting approximately 2 mg Cd/L solution was prepared; 1.569 mg Cd/L was later recovered in ICP-OES (Spectro Genesis, Germany). The initial pH was adjusted to 7 for the feed solution. 50 ml solution was put into a centrifuge tube and 1 g (@ 2% w/v) OA added to it. After 2 hours' shaking at 200 rpm the mixtures were filtered with ash-less filter paper (Whatman no. 42). Cd<sup>2+</sup> concentration of the aliquots was determined by ICP-OES.

Each amendment was tested in five replications. Using the following equation the percentage Cd removed from the aqueous solution was calculated (Cheraghi et al. 2015):

$$\%Cd\text{ removal} = \frac{(C_0 - C_e)}{C_0} \times 100$$

Where,  $C_0$  = Initial Cd<sup>2+</sup> concentration (mg/L) and  $C_e$  = Cd<sup>2+</sup> concentration after adsorption (mg/L).

**Table 2** Cadmium content and CEC of the organic amendments

Organic materials	Source	Total Cd (mg/kg)	Bioavailable Cd (mg/kg)	CEC (cmol/kg)
Cow dung	Collected from nearby farm	2.35	0.001	219
MOC	Collected from nearby oil mill	0.05	0.001	230
Rice husk	Collected from nearby rice mill	0.05	0.001	226
Saw dust	Collected from nearby saw mill	0.05	0.001	171
Tea leaf	Collected from tea stalls	0.05	0.001	310
Vermicompost	Purchase from market	0.05	0.001	68

Note: MOC (Mustard oil cake), CEC (Cation exchange capacity)

The study was conducted under open conditions in a net house at Khulna University campus (22.802° N, 89.533° E). Earthen pots (40 cm height and 30 cm diameter) were prepared for taking 5 kg soil. 100 g of each OA were added with 5 kg soil to obtain a 2% rate (40 t OA/ha soil) of amendment, while 200 g was added to get a 4% rate (80 t OA/ha soil) of amendment. Added OA were uniformly mixed with soil. Half of the prepared pots were spiked with Cd (10 mg Cd/kg soil). Standard Cd solution (1000 mg/L) was prepared by dissolving CdCl<sub>2</sub>.H<sub>2</sub>O (Sigma-Aldrich, Germany). 25 ml standard Cd solution was diluted up to 2 L by adding water. Half of the soil was poured into the pot and then the prepared Cd solution was added. Finally, the remaining soil was instantly added to ensure homogenous Cd<sup>2+</sup> distribution in the soil. Field capacity water was maintained initially (21 days) and then the soil was kept submerged at 3–4 cm (7 days).

The crop was grown under pot conditions in both agricultural soil (without Cd stress) and Cd-stressed soil (10 mg/kg). Two vigorous seedlings that were four weeks old were transplanted into each pot. Saturated water was maintained throughout the experiment till a week before harvest.

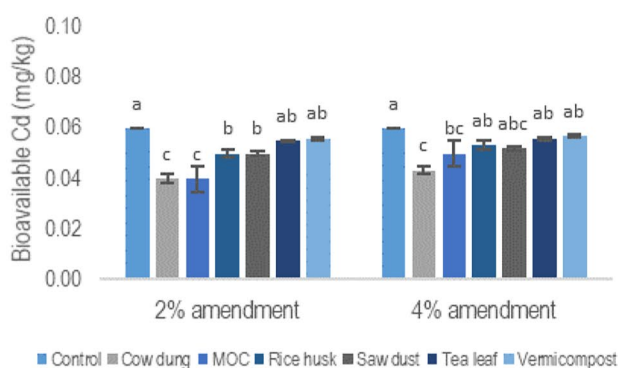
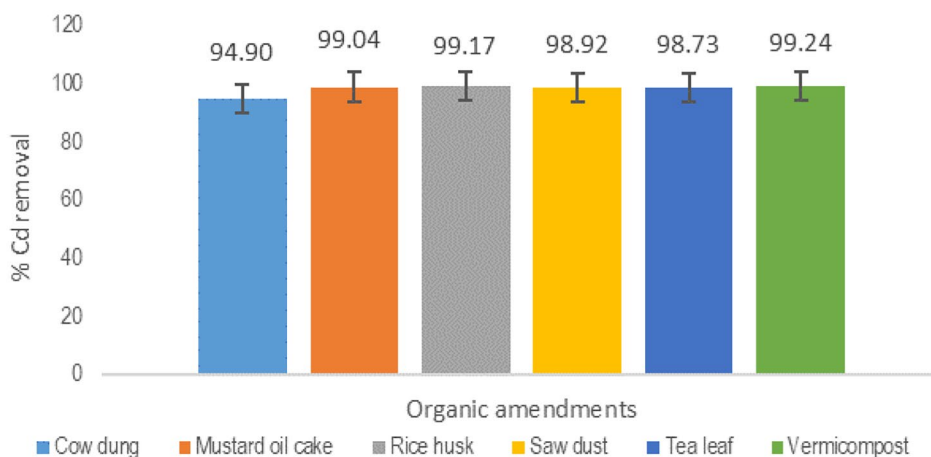
Recommended doses of fertilizers (urea 75, TSP 150, muriate of potash 115 and gypsum 150 kg/ha) and pesticides were applied. In total, 78 pots were arranged in a randomized complete block design to reduce experimental errors.

Rice grains were sampled at ripening and washed twice with distilled water. Then these grains were oven dried at 65° C and the chaff of the grain was manually separated. The brown rice samples were preserved for analysis. Rice grains were crushed and ground to homogenize the sample. Soil sampled from experimental pots was air dried, smashed and sieved (2 mm) for Cd analysis.

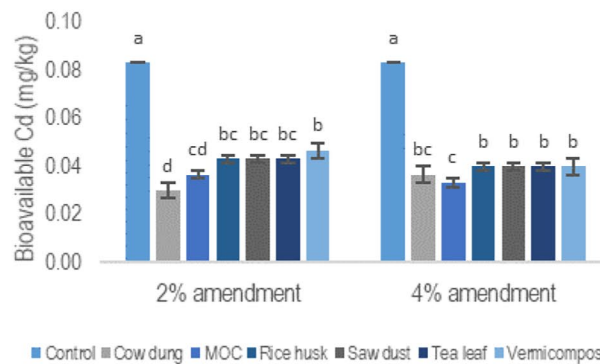
Hydrometer method (Gilson, USA) served to determine particle size (Gee and Bauder 1986). Soil pH was measured by pH meter (Hanna HI110, USA) using distilled water at a 1:2.5 ratio (Li et al. 2005). EC was measured by EC meter (Hanna HI2315, USA) in 1:5 ratio (Hardie and Doyle 2012). Organic matter analysis was conducted through titration as described by Nelson and Sommers (2001). CEC was determined by using NH<sub>4</sub>OAc (Lu 2000). Bioavailable soil Cd was determined by utilizing CaCl<sub>2</sub> solution (0.01 M) at a ratio of 1:10 after shaking that lasted 2 h (Houba et al. 2000). Grain and soil samples were digested with HNO<sub>3</sub>-HClO<sub>4</sub> acid mixture (2:1 ratio) following the method of Hseu (2004) for total analysis of Cd. ICP-OES (Spectro Genesis, Germany) measured the Cd<sup>2+</sup> concentrations. Sequential extraction as described by Tsai et al. (2003) was employed to determine soil Cd fractions.

Bioconcentration factor (BCF) is calculated according to the following equation (Jiale et al. 2021):

**Fig. 1** Percent removal of Cd from solution by organic amendments



a) Agricultural soil



b) Cd stress soil

**Fig. 2** Soil bioavailable Cd as influenced by six organic amendments. Lower case letters at the top of the bars represents differences among individual means as compared by using LSD test at 0.05 level of significance

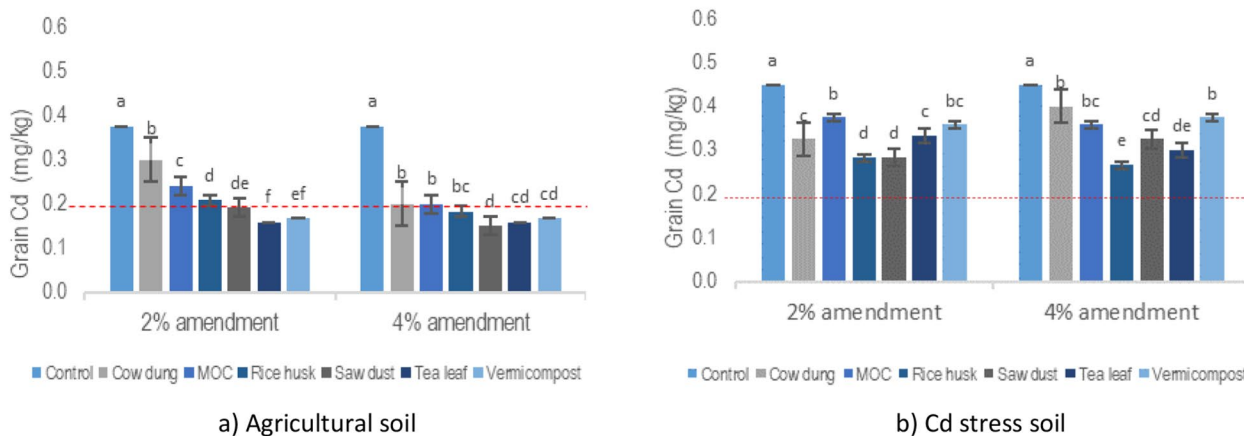
$$BCF = \frac{\text{Grain Cd}}{\text{Soil bioavailable Cd}}$$

Grade-1 water of 0.01 mS/m EC (at 25° C) was used for laboratory analysis. Standard solution purchased from Sigma-Aldrich, Germany (Multi-element IV, SKU- 1,113,550,100) was used to calibrate ICP. Reference rice flour certified material for trace elements (SKU- IRMM804-15G, Sigma-Aldrich, Germany) was used from which 94.5–96.0% Cd was recovered. Reagent blank, replicated samples, triple reading and continuing check verifications (CCVs) were implemented while readings were taken in ICP for each sample batch.

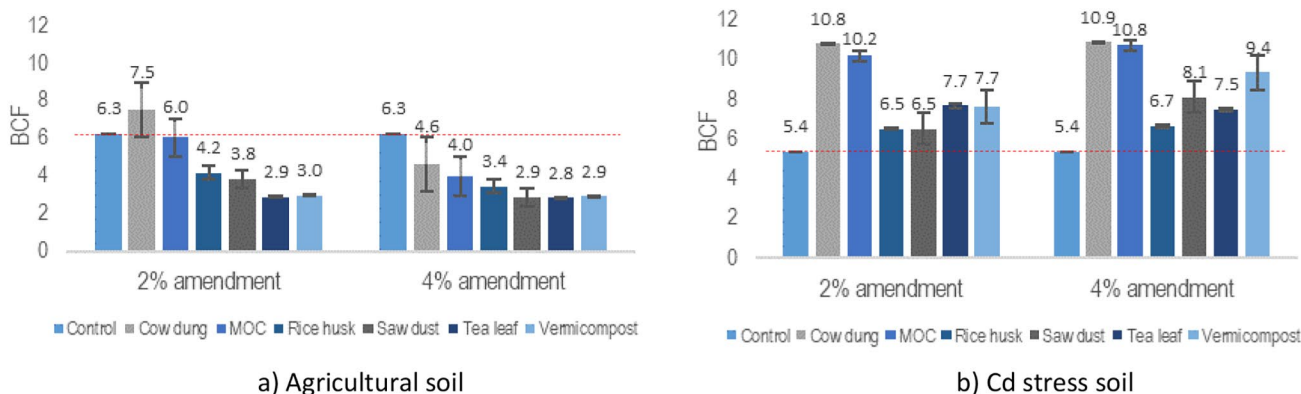
Statistix 10 software was used for data analysis. One-way ANOVA (analysis of variance) at confidence intervals of 95% ( $P < 0.05$ ) served for the statistical analysis. Where significant, LSD were calculated among the multiple comparisons of means using Tukey multiple range test. When  $P < 0.05$  then the treatment effect was statistically significant.

## Results and Discussion

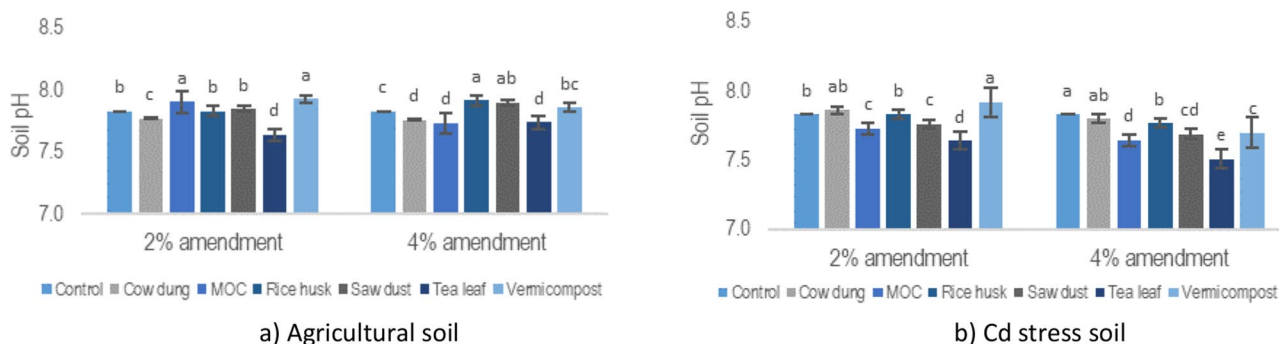
All six OAs were found to be very efficient in removing Cd from aqueous solution at 2% application rate (Fig. 1). Except for cow dung the other five OAs adsorbed more than 99% solution  $\text{Cd}^{2+}$  ions. Cow dung removed 95% Cd from the solution. Therefore, OAs are potential agents to adsorb bioavailable Cd from soil solution. Low-cost OAs obtained from animal wastes and agricultural sources are of prime interest for this process (Mathew et al. 2016). Deploying low-cost OAs helps to reduce environmental pollution and minimize production costs. Agricultural use of these OM adds nutrients to soil through decomposition and this also helps to reduce environmental pollution or damage done to the soils. Effective Cd removal from aqueous solution using various biosorbents was reported by several authors (Cheraghi et al. 2015; Tatah et al. 2017; Alalwan et al. 2020; Visa et al. 2020). However, Mathew et al. (2016) argued that the adsorption process depends on several parameters,



**Fig. 3** Grain Cd content of SL-8 rice varieties at different organic amended agricultural soil. Red dotted lines indicate the permissible limit for Cd in rice grain (0.2 mg/kg)



**Fig. 4** BCF of rice grain as influenced by organic amendments and its rate of application. Red lines indicate the BCF value of control



**Fig. 5** Soil pH after harvest of six organic amended soil

especially temperature, pH, pressure, adsorbent activation and adsorbent surface area.

Agricultural and Cd-stressed soil (10 mg Cd/kg soil) contained 0.06 and 0.08 mg/kg Cd in control pots (without OA), respectively. The salt CdCl<sub>2</sub>·H<sub>2</sub>O added Cd in soil as Cd<sup>2+</sup> which increased its bioavailability by about 38%. The effects of OA on Cd bioavailability in agricultural and

Cd-stressed soil are presented in Fig. 2a and b. In both cases, the addition of OA noticeably reduced bioavailability of Cd. At a 2% rate of application, cow dung proved to be most effective and reduced Cd bioavailability by about 33% in agricultural soil and about 64% in Cd-stressed soil. MOC also diminished the Cd concentration in bioavailable pool remarkably well. The performance of rice husk, saw



dust and tea leaf was very similar. Among the six amendments VC was found to be the least efficient in adsorbing Cd, achieving only 7% in agricultural soil and 43% in Cd-stressed soil compared with the control. Reduction of Cd bioavailability due to OA application was reported in other research (Bolan et al. 2003; Ruttens et al. 2006; Liu et al. 2009; Ok et al. 2011; Park et al. 2011; Wu et al. 2011; Juang et al. 2011). In the case of 4% application, rate the removal of Cd from the soil solution did not increase considerably compared to the 2% application rate. Therefore, up to 2% application of OA apparently reduces soil bioavailable Cd.

Rice grain Cd content grown under different OA treated soils is presented in Fig. 3a and b. Addition of OA in soil noticeably reduced grain Cd content irrespective of soil and rate of application compared with the control. In agricultural soil, grain Cd content grown in control pots were 0.38 mg/kg and for OA-added pots it was in the 0.15 to 0.30 mg/kg range. Saw dust, tea leaf and VC produced grains with safe amounts of Cd in this soil that made human consumption secure (<0.2 mg/kg). Cow dung and MOC apparently lowered grain Cd with an increase in the application rate from 2% to 4%. Mamun et al. (2021) recently recommended 1% VC application as the effective dose to reduce Cd uptake from soils in Bangladesh. Cd content in grains grown in the control Cd-stressed soil amounted to 0.45 mg/kg while OA-added soils it was in the 0.27 to 0.40 mg/kg range. 4% OA application rate was not even enough to lower grain Cd below the safe value. In both application rates, rice husk proved to be the most efficient in reducing grain Cd. Other OAs showed significant variation in lowering grain Cd. Many researchers recommended the use of various OAs to reduce Cd toxicity to plants (Rizwan et al. 2016a). The response of plants to Cd concentration considerably varied according to the type of soil and OA application rate (Rizwan et al. 2016b). Liu et al. (2020) stated that reduced Cd uptake by rice is possibly due to the influence of OA on soil properties to reduce Cd bioavailability.

BCF of SL-8 rice variety considerably varied due to the application of OA compared with the control irrespective of soil type and application rate (Fig. 4a and b). In agricultural soil, BCF value ranges from 2.8 to as high as 7.5. In most instances, OA application in both 2% and 4% rates noticeably reduced BCF. Rice husk, saw dust, tea leaf and VC efficiently reduced BCF value at the 2% rate of application whereas cow dung and MOC were efficient at the 4% rate. However, in Cd-stressed soil OA application raised the BCF value higher than that of the control (Fig. 4b). The lowest BCF was found in the control (5.4) and the highest value (10.9) was recorded in rice grains grown in cow dung-treated soils. Again cow dung and MOC emerged as the least effective. Rate of OA application did not show remarkable variation in Cd-stressed soil. Though both bioavailable

Cd in soil and grain Cd were reduced by the OA addition the Cd uptake by SL-8 rice variety markedly increased due to Cd spiking in soil. The Cd BCF of rice is significantly higher than wheat and maize (Chen et al. 2021). The authors also reported that soil Cd content, pH, OA and other factors influence Cd BCF of rice.

All OAs greatly influenced the pH of soils (Fig. 5a and b). In agricultural soil, pH variation was in the range of 7.64 to 7.93 in 2% and 7.74 to 7.92 in 4% application rate. In contrast, pH variation in Cd-stressed soil was 7.64 to 7.92 at 2% and 7.64 to 7.83 at 4% OA application. Some amendments increased soil pH whereas others curtailed it. Soil pH lowering was noticeably observed in Cd-stressed soil at 4% application rate. With the increase in OA application rate organic acids are produced through decomposition which reduces soil pH and increases bioavailable Cd in soil (Beesley et al. 2010; Jiang et al. 2012). Moreover, organic acids present in rhizosphere soil can lower the pH value and elevate the bioavailability of heavy metals by activating insoluble forms (Zhi et al. 2020). Therefore, Cd chemistry in soil to a large extent depends on the variation in soil pH. Jiale et al. (2021) reported there was a negative correlation between bioavailable Cd and soil pH. The Authors also stated that the risk of Cd toxicity cannot be neglected specially upon soil acidification which elevates Cd bioavailability and encourages Cd deposition in rice grain. Hence, considering the solubility of Cd with soil pH the application rate of the OA should be judged carefully.

The influence of OA on soil pH also varies with time dynamics after application in soil (Shi et al. 2011). Significant elevation in soil pH due to the addition of crop straw and compost was reported in other studies (Lombi et al. 2003; Zhu et al. 2012; Wang et al. 2015; Sun et al. 2020). Meng et al. (2019) reported that Cd bioavailability may also be effectively reduced by increasing soil pH. The authors showed through linear regression analysis that elevating soil pH bears a positive correlation to immobilizing Cd from soil solution. The elevation in soil pH increases the amount of negative charge on various soil colloidal constituents which enhances Cd sorption to reduce its bioavailability to plants (Naidu et al. 1994; Bolan et al. 2014). The ratio of total soil Cd and soil solution Cd is defined as solid-liquid distribution coefficient ( $K_d$ ) of Cd (Degryse et al. 2009). The coefficient is mainly soil pH-dependent (Degryse et al. 2009) and Alloway (2013) convincingly showed that each unit pH rise causes an increase in  $K_d$  by about a factor of four.

## Conclusion

Six traditional OAs used in this study successfully removed more the 95% Cd from aqueous solution. In both agricultural and Cd-stressed soils the addition of OA markedly reduced Cd bioavailability. Addition of OA in soil remarkably reduced grain Cd content irrespective of soil and rate of application compared with the control sample. Saw dust, tea leaf and VC produced grains with safe amounts of Cd for humans to consume (<0.2 mg/kg). Tea leaf was the most efficient OA for minimizing rice grain Cd in agricultural soil and rice husk appeared to be the most efficient at reducing grain Cd in Cd-stressed soil. 2% (40 t/ha) application of OA apparently reduced Cd bioavailability in soil. BCF of SL-8 rice variety considerably varied due to the application of OA compared with the control irrespective of soil type and application rate. Despite the fact that both bioavailable Cd in soil and grain Cd were reduced by OA addition, the SL-8 rice variety's uptake of Cd markedly increased as a result of Cd spiking in soil. All OAs clearly had an effect on soil pH which bears close relationship with Cd availability and plant growth. Hence, agrochemicals should be carefully judged before applying them to soil. Lastly, the addition of OA in soil makes the dynamic equilibrium very complicated so more detailed studies are required to evaluate its role in regulating Cd availability in rhizosphere and its subsequent deposition in rice grain.

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**Data Availability** All data are available.

**Code Availability** Not applicable.

## Declarations

**Conflicts of Interest** The authors declare that they have no conflict of interest.

All sections are relevant to the manuscript.

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