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Review

Effects of natural organic matter on cadmium mobility in paddy soil: A review

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ABSTRACT

Cadmium (Cd) contamination in paddy soil has caused public concern. The uptake of Cd by rice plants depends on soil Cd mobility, which is in turn substantially influenced by organic matter (OM). In this review, we first summarize the fate of Cd in soil and the role of OM. We then focus on the effects of OM on Cd mobility in paddy soil and the factors influencing the remedial effectiveness of OM amendments. We further discuss the performance of straw incorporation in the remediation of Cd-contaminated paddy soils reported in laboratory and field studies. Considering the huge production of organic materials (such as straw) in agriculture, the use of natural OM for soil remediation has obvious appeal due to the environmental benefits and low cost. Although there have been successful application cases, the properties of OM amendments and soil can significantly affect the remedial performance of the OM amendments. Importantly, straw incorporation alone does not often decrease the mobility of Cd in soil or the Cd content in rice grains. Careful evaluation is required when considering natural OM amendments, and the factors and mechanisms that influence their remedial effectiveness need further investigation in paddy soil with realistic Cd concentrations.

Introduction

Cadmium (Cd) is a toxic metal that can cause diseases such as cancers, renal diseases, osteomalacia, and osteoporosis (Maret and Moulis, 2013). Rice is the major exposure route for Cd for people with rice as their staple food. For example, rice contributes 56% of the dietary Cd exposure for Chinese people (Song et al., 2017). Japanese people also obtain about one-half of their Cd from rice (Suda and Makino, 2016). Rice accumulates Cd from contaminated soils more efficiently than other crops (Maret and Moulis, 2013), and polished rice samples contaminated by Cd (containing > 200 μg/kg Cd; the European and Chinese standards) have been reported in many markets in the world (Shi et al., 2020). In 2008, researchers found that about 10% of the 91 rice samples from Chinese markets contained > 200 μg/kg Cd (Zhen et al., 2008). After ten years, the exceedance rate has not decreased. In 2018,

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Chen et al. (2018) collected 160 polished rice samples from Chinese markets and still detected 10% of samples containing > 200 μg/kg Cd.

The uptake of Cd by rice depends on the mobility of Cd in soil, which is in turn substantially influenced by organic matter (OM). Mobile Cd generally includes dissolved and weakly adsorbed Cd. OM can effectively adsorb Cd due to the presence of functional groups (e.g., hydroxyl, carboxyl, and phenolic groups) (Smolders and Mertens, 2013). The addition of OM to soil may increase the Cd adsorption capacity of the soil (Yuan et al., 2019). Moreover, OM addition can directly or indirectly alter soil pH (Yuan et al., 2019), which is the key factor influencing Cd partitioning in soil (Smolders and Mertens, 2013). Being cost-effective and environment-friendly, various natural (e.g., plant residues and animal manures) and processed (e.g., biochar) organic materials have been used to remediate Cd-contaminated soils, which has been reviewed recently (Hamid et al., 2020; Khan et al., 2017).

Previous reviews provided general information on the application of OM in Cd-contaminated soils, but they did not focus on paddy soil and paid little attention to the influence of OM on the chemical and microbial processes that regulate soil Cd speciation and mobility. In this review, we first summarize the fate of Cd in soil and the role of OM. We then focus on the effects of OM on Cd mobility in paddy soil and the factors influencing the remedial effectiveness of OM amendments. We further discuss the performance of straw incorporation, a common practice in paddy fields, in the remediation of Cd-contaminated paddy soils as reported in laboratory and field experiments. The scope of this review is limited to natural OM. For the application of biochar, the audience may refer to Hu et al. (2016), O'Connor et al. (2018), and Hamid et al. (2020). We hope that this review will help us better understand the remedial effects of natural OM and better manage Cd-contaminated paddy soils.

1. Cd contamination of paddy soil

Paddy soils contaminated by Cd have been reported in many rice-producing countries (Chunhabundit, 2016; Khanam et al., 2020; Liu et al., 2016) (Table 1). Industry activities such as mining and smelting operations are the main sources of soil Cd contamination (Khaokaew et al., 2016). Fertilizers and agrochemicals, irrigation with contaminated water, and natural sources can also result in elevated soil Cd concentrations (Table 1). Moreover, the application of some Cd-containing organic materials such sewage sludge and livestock manures will bring Cd to soil (Hamid et al., 2020; Khan et al., 2017). Liu et al. (2016) surveyed 187 administrative regions with rice cultivation in China and found Cd-contaminated paddy soils in 1/3 of the surveyed regions, mainly in Hunan, Jiangxi, Guangxi, Guangdong, Chongqing, and eastern Sichuan. However, the specific area of Cd-contaminated rice fields in China remains unknown. Liu et al. (2016) identified the sources of Cd contamination for 15% of the surveyed regions. Pollution sources included mining and metallurgical industries, other industries, sewage irrigation, and electronic waste, with mining activities being the main source.

High soil Cd concentrations often, although not always, cause Cd contamination of rice (Table 1). To protect public health, many countries have issued standards or regulations regarding the maximum concentrations of Cd allowed in soil and rice. An allowable limit of 0.4 or 0.2 mg Cd per kg rice is adopted by many countries (Table 2) and the European Union (Shi et al., 2020). Correspondingly, the total Cd concentration should be below 1.5–6 mg/kg in agricultural soils (in China, India, and Vietnam) or below 0.01 mg/L in soil leachate (in Japan) (Table 2). Some countries have not set standards for allowable Cd concentrations in soil or rice, and therefore international standards or the standards from other countries are used in the literature (see references in Table 2).

2. Fate of Cd in soil and the role of OM

The Cd concentration in soil solutions is generally very low, being < 0.1–20 μg/L in uncontaminated soils and 0.4–200 μg/L in contaminated soils (Smolders and Mertens, 2013). Organic and inorganic ligands in soil solutions, such as dissolved organic matter (DOM) and chloride ions, can form complexes with Cd, thereby promoting the dissolution of Cd (Smolders and Mertens, 2013).

Most soil Cd is adsorbed to OM, oxyhydroxides of Fe, Al, and Mn, and clay minerals, among which clay minerals generally make a small contribution to Cd adsorption (Sauvé et al., 2000b; Smolders and Mertens, 2013). In flooded paddy soil, Cd may also precipitate as cadmium carbonates when the soil is alkaline and the Cd content is high (Khaokaew et al., 2011), in secondary iron minerals (e.g., magnetite) when the soil iron content is high (Muehe et al., 2013a), or as CdS when the soil contains enough sulfur (Fulda, et al., 2013b). The mechanism of Cd adsorption is that Cd^{2+} binds to the surface oxygen atoms of the hydroxyl, carboxyl, or phenolic groups of humus or oxyhydroxides through inner sphere adsorption (Smolders and Mertens, 2013):

$$S\text{–}OH + Cd^{2+} \rightarrow S\text{–}OCd^{+} + H^{+} \quad (1)$$

Although the adsorption of Cd by soil can reach equilibrium within one hour (Christensen, 1984a), it is almost completely reversible even after one year (Christensen, 1984b). This reversibility may be because the ionic radius of Cd^{2+} is much larger than that of Fe^{3+} or Al^{3+}, so it is difficult for Cd to enter the lattice of Fe or Al oxides by isomorphic substitution (Smolders and Mertens, 2013). Synthesis experiments indeed show that Cd has difficulty replacing Fe in lepidocrocite, a common iron oxide in soil (Gräfe and Singh, 2006). Therefore, aging scarcely leads to the fixation of Cd, and most of the Cd in soil can be easily released (Smolders and Degryse, 2006; Smolders and Mertens, 2013).

The key factor affecting Cd adsorption by soil is pH. It can be seen from Eq. (1) that H^{+} is the main competing cation for Cd^{2+} adsorption on OM or oxyhydroxides. Studies have shown that if soil pH is increased by one unit, the adsorption of Cd by soil will increase by 3–5 times (Christensen, 1984a; Degryse et al., 2009; Sauvé et al., 2000a). The effect of soil texture on Cd adsorption is weak (Christensen, 1984a).

Table 1 – Cases of Cd contamination of paddy soil in major rice-producing countries.

Country	Site	Total soil Cd (mg/kg)	Rice grain Cd (mg/kg)	Cause	Reference
Bangladesh	Dhaka	0.1–1.8	NA	Industries	Kashem and Singh (1999)
China	Lianhuashan tungsten mine area	0.22–1.6	0.04–0.33	Mining	Yu et al. (2016b)
India	Ropar wetland area	0.31–4.60	0.57–1.12	Atmospheric deposition of fly ash released from factories; application of pesticides	Sharma et al. (2018)
Japan	Fukui	0.2–10.4	0.02–1.82	Mining	Takijima and Katsumi (1973)
Malaysia	Kedah; Penang	3.54–20.86	NA	High background value; fertilizers and pesticides	Jamil et al. (2011)
Philippines	Naboc River area	< 1	0.057–1.025	Irrigation using contaminated river water	Appleton et al. (2006)
South Korea	Daduk mine area	0.40–4.76	0.03–0.65	Mining	Lee et al. (2001)
Thailand	Phatat Pha Daeng	0.5–284	0.05–7.7	Mining	Simmons et al. (2005)
Vietnam	Tan Long	2.3–42.9	NA	Mining	Chu (2011)

NA: not available.

Table 2 – Allowable limits for Cd in rice and agricultural soils in major rice-producing countries.

Country	Allowable limit for Cd in agricultural soils (mg/kg)	Reference	Allowable limit for Cd in rice (mg/kg)	Reference
Bangladesh	NE or NA[a]	Islam et al. (2018)	NE or NA [a]	Islam et al. (2018)
China	1.5–4[b]	China Ministry of Ecology and Environment (2018)	0.2	China National Health Commission and National Medical Products Administration (2017)
India	3–6	Kumar Sharma et al. (2007)	0.4	Food Safety and Standards Authority of India (2011)
Japan	Soil concentration ≤ 150 mg/kg; Soil leachate ≤ 0.01 mg/L	Makino et al. (2019)	0.4	Makino et al. (2019)
Malaysia	NE	Najib et al. (2012)	0.4	Malaysia Food Regulations (1985)
Philippines	NE or NA	Appleton et al. (2006)	NE or NA	Abratique Layosa et al. (2018)
South Korea	NE	Antoniadis et al. (2019)	0.2	Ahn et al. (2017)
Thailand	0.15[c]	Chaiwonga et al. (2013)	NE	Kerdthep et al. (2009)
Vietnam	2	Chaiwonga et al. (2013)	0.4	Phan et al. (2016)

[a] NE: non-existent; NA: not available.
[b] Risk intervention value, depending on soil pH.
[c] Investigation level.

The concentration of dissolved Cd in soil can be predicted by some empirical models (Degryse et al., 2009; Sauvé et al., 2000a, 2000b). These models use linear regression to investigate the relationship of the dissolved metal concentration or the distribution coefficient K_d with soil pH alone or with soil pH and total organic carbon (TOC). K_d is the ratio of metal concentration in the solid phase (mg/kg) to that in the solution phase (mg/L); metal concentration in the solid phase is sometimes substituted by total metal concentration in soil (Degryse et al., 2009; Sauvé et al., 2000a, 2000b). pH plays a major role in predicting the K_d of Cd, and the inclusion of TOC will improve the model. For example, Sauvé et al. (2000a) analyzed the data from over 70 different studies, and obtained the following relationship for soil Cd:

$$\log_{10} K_d = -0.60 + 0.49 \times \text{pH} \ (n = 830, R^2 = 0.467, p < 0.001) \quad (2)$$

$$\log_{10} K_d = -0.65 + 0.48 \times \text{pH} + 0.82 \times \log_{10}\text{TOC} \ (n = 751, R^2 = 0.613, p < 0.001) \quad (3)$$

$$\log_{10} (\text{dissolved Cd concentration}) = 3.42 - 0.47 \times \text{pH} + 1.08 \times \log_{10} (\text{total Cd concentration}) - 0.81 \times \log_{10}\text{TOC} \ (n = 751, R^2 = 0.884, p < 0.001) \quad (4)$$

However, the application of these models in paddy soil has not been reported.

3. Effects of OM addition on Cd mobility in paddy soil

OM is essential for maintaining soil fertility and ecological functions (Chapin et al., 2011; Weil and Brady, 2016). Various organic materials have been used to increase soil OM content, such as farmyard manure, animal wastes, crop residues, commercial composts, green manures, night soil, sewage, sludge, and organic wastes from industry (Hesse, 1984; Wen, 1984). Most of these organic materials have been tested for the remediation of Cd-contaminated soils (Hamid et al., 2020; Khan et al., 2017).

3.1. *Direct and indirect effects of OM on soil Cd mobility*

Adding OM will directly or indirectly influence Cd mobility in paddy soil. Directly, as mentioned above, OM can adsorb Cd. Indirectly, OM can adjust soil pH and therefore alter Cd adsorption by soil. Under anaerobic conditions, OM can also regulate microbial reduction processes in soil and promote the precipitation of Cd (such as the formation of CdS).

The influence of OM on soil pH also has direct and indirect mechanisms. There are three direct mechanisms. First, the added OM may contain acidity or alkalinity. The pH of most animal wastes is neutral to alkaline and of green manures and straws neutral to acid (**Appendix A Table S1**). When the pH of the added OM is higher than that of soil, OM addition is expected to increase soil pH, and *vice versa*. Second, adding OM will enhance the pH buffering capacity of soil (Yuan et al., 2016). Lastly, during OM decomposition, the release of CO_2 and organic acids (Weil and Brady, 2016) will lower soil pH, while the generation of NH_4^+ (Weil and Brady, 2016) and decarboxylation may increase soil pH (Yan and Schubert, 2000; Yan et al., 1996):

$$\text{R-NH}_2 + H_2O + H^+ \rightarrow \text{R-OH} + NH_4^+ \quad (5)$$

$$\text{R-COO}^- + H^+ \rightarrow \text{R-H} + CO_2 \quad (6)$$

Indirectly, as a carbon source and electron donor, OM can promote the microbe-mediated reduction reactions in soil under anaerobic conditions. After flooding, oxygen is quickly depleted, and NO_3^-, Mn(IV), Fe(III), and SO_4^{2-} are then sequentially reduced to N_2/NH_4^+, Mn(II), Fe(II), and H_2S under the action of microorganisms (Borch et al., 2009; Ponnamperuma, 1972; Reddy and DeLaune, 2008). These reduction reactions consume protons, and the resulting increase in soil pH will promote Cd adsorption. As pH is the prime factor regulating Cd adsorption by soil, when Cd is immobilized after adding OM to flooded soil, it is mainly attributed to the increase in soil pH rather than Cd adsorption by the added OM (Chen, 2013; Yuan et al., 2019).

3.2. *The effects of iron and sulfur redox reactions on soil Cd mobility and the role of OM*

The effects of iron and sulfur redox reactions on Cd mobility are of particular concern (Fulda, et al., 2013b; Yu et al., 2016a; Zhang et al., 2012). After flooding, the reductive dissolution of iron oxide will release the Cd adsorbed on it (Muehe et al., 2013a; Yu et al., 2016b); however, the secondary iron minerals formed during iron reduction can immobilize Cd through adsorption and co-precipitation (Li et al., 2016; Muehe et al., 2013a, 2013b) (**Fig. 1**). Muehe et al. (2013a) reported that in an iron-rich soil, iron reduction after flooding caused Cd immobilization by a secondary iron mineral (probably magnetite), thereby reducing soil Cd mobility. Similarly, Li et al. (2016) studied the fate of Cd during the microbial reduction of Cd-containing polyferric flocs in bioreactors, and they found that iron reduction first caused the release of Cd, but with the generation of secondary iron minerals (goethite and magnetite) Cd was adsorbed again.

Cd can react with H_2S, the product of sulfate reduction, to form insoluble cadmium sulfide precipitates, lowering soil Cd mobility after flooding (Dvorak et al., 1992; Jiang and Fan, 2008; Ponnamperuma, 1972) (**Fig. 1**). Pure CdS formation is unlikely to occur in soil; Cd^{2+} generally substitutes into other common metal sulfides such as FeS (Barrett and McBride, 2007). It should be noted that when the concentration of sulfate (and thus the S^{2-} generated by sulfate reduction) is limited, the precipitation of other metal sulfides, which also consumes S^{2-}, will make the precipitation of CdS difficult (de Livera et al., 2011; Fulda et al., 2013a, 2013b). Thermodynamic equilibrium models predict that when multiple metal ion species coexist with sulfur ions, metal sulfides with lower solubility products will precipitate first (Weber et al., 2009a). For example, the solubility product of CuS ($K_{sp}{}^0 = 10^{-22.3}$) is about 8 orders of magnitude lower than that of CdS ($K_{sp}{}^0 = 10^{-14.4}$), so when Cd^{2+}, Cu^{2+}, and S^{2-} coexist, CuS will be formed first; when Cu^{2+} is almost depleted, CdS will be then formed (Weber et al., 2009a). Nevertheless, this sequential mode can be affected by the kinetics of reactions such as desorption, diffusion, and precipitation (Weber et al., 2009a). Metal sulfides can adsorb metal ions, further facilitating Cd immobilization (Jong and Parry, 2004; Watson et al., 1995). However, some studies have shown that metal sulfide colloids formed after flooding may increase metal mobility in the short term (Abgottspon et al., 2015; Weber et al., 2009b).

Most of the microorganisms involved in iron and sulfate reduction are heterotrophic and require OM to provide energy and carbon (Lovley, 1991; Muyzer and Stams, 2008). If there is a lack of OM, the reduction of iron and sulfate after soil flooding may be impeded (Yuan et al., 2015a, 2015b), and the resulting immobilization of Cd may also be hindered. Conversely, if OM is added to flooded soil, it may promote iron and sulfate reduction and Cd immobilization (de Livera et al., 2011; Yuan et al., 2019). Therefore, adding OM after flooding is a potential measure to decrease Cd mobility in paddy soils (de Livera et al., 2011; Jiang and Fan, 2008; Li et al., 2017; Yuan et al., 2019).

After drainage, H^+ generated during iron and sulfur oxidation will compete with Cd^{2+} for adsorption sites, resulting in the release of adsorbed Cd (Fulda, et al., 2013b; Li et al., 2017; Smolders and Mertens, 2013). In addition, the oxidative dissolution of CdS will release Cd (de Livera et al., 2011; Fulda, et al., 2013b) (**Fig. 1**). Different metal sulfides have different oxidation rates, and FeS oxidizes faster than CdS, CuS, PbS, and ZnS (Simpson et al., 2000). If a large amount of FeS has been formed under reducing conditions, the oxidation of CdS, CuS, PbS, and ZnS after drainage may be retarded, because FeS will be preferentially oxidized (Tai et al., 2017). In paddy soil suspensions, de Livera et al. (2011) reported that due to the enhanced metal sulfide precipitation under reducing conditions, the dissolved Cd concentration that increased after short-term soil oxidation was still lower than the level before soil reduction.

OM can buffer the decrease in soil pH (Paul and Ulf, 2011). OM can adsorb Cd. In addition, OM decomposition after drainage will consume oxygen, and OM can coat metal sulfides, so it has an inhibitory effect on the oxidation of sulfides (Bush and Sullivan, 1999; Rigby et al., 2006; Yuan et al., 2016).

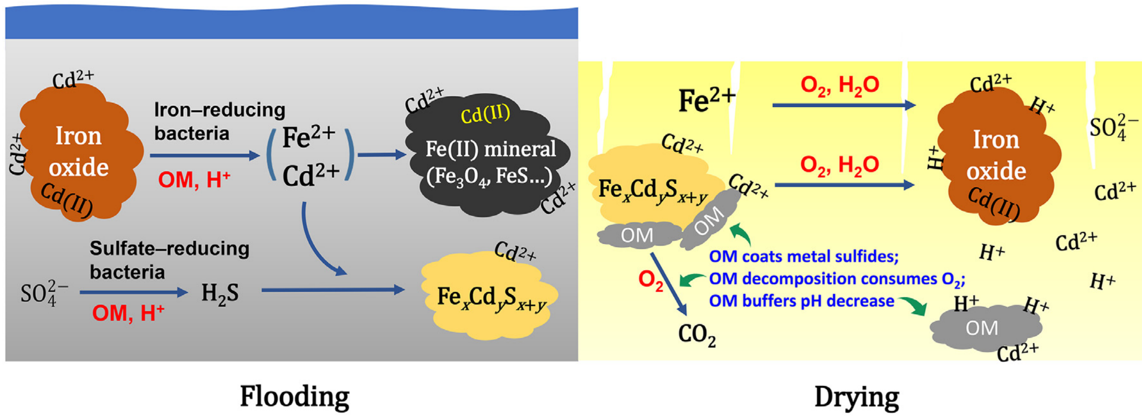

Fig. 1 – Proposed effects of iron and sulfur redox reactions on Cd mobility in paddy soil. After flooding, the reductive dissolution of iron oxide will release adsorbed Cd, but secondary iron minerals formed may immobilize Cd. Cd can also precipitate as CdS under sulfate-reducing conditions. After drying, soil oxidation results in lowered pH, promoting Cd desorption. The oxidation of CdS will also release Cd. OM may retard Cd mobilization after soil drying because OM can buffer the decrease in soil pH, OM decomposition will consume oxygen, and OM can coat metal sulfides, but this requires further investigation.

Therefore, the presence of OM may slow the Cd mobilization after soil drainage, which is worth further investigation (Fig. 1).

Recently, Yuan et al. (2019) investigated the change in Cd solubility with time after flooding and drainage in paddy soil microcosms. The authors found that the addition of 20 g/kg rice straw efficiently promoted microbial iron reduction after soil flooding, which resulted in a significant increase in soil pH and immobilization of Cd. The decrease in dissolved Cd was unlikely to be caused by secondary iron minerals. The addition of 20 g/kg hematite (a common iron oxide in paddy soil) or 300 mg/kg gypsum-S was not effective. After five weeks of drainage and aeration, the concentration of dissolved soil Cd increased but did not reach the level before flooding, and the concentration of dissolved Cd in the straw-amended soil was still the lowest among treatments. The authors concluded that OM can efficiently promote microbial reduction processes and Cd immobilization in paddy soil after flooding. The addition of iron oxide or sulfate alone is ineffective, because the limitation of electron donors on microbial activities cannot be eliminated.

3.3. *Cd adsorption by microbial cells*

It is worth noting that, as a part of soil OM, microbial cells also have colloidal properties and can increase the surface area and adsorption capacity of soil (Dickinson, 2015; Ha et al., 2010; Yu and Fein, 2015). Many studies in cell suspensions indicate that due to the existence of various functional groups (such as carboxyl, sulfhydryl, phosphoryl, and amide groups) on the cell wall, microbial cells can adsorb metal ions, including Cd^{2+} (Chang et al., 1997; Ha et al., 2010; Kaulbach et al., 2005; Mishra et al., 2010; Yu and Fein, 2015). In a bioreactor experiment, Yuan et al. (2018) reported that 422 µg/L of dissolved Cd completely transferred to adsorbed Cd during microbial iron reduction, and the cells of the iron-reducing bacteria probably contributed to 2/3 of the Cd adsorption. However, to estimate the quantity of Cd adsorbed on microbial cells in a real soil could be challenging and has not been reported yet.

Using ultraviolet irradiation, Sun and coworkers obtained a mutant (B38) of the bacterium *Bacillus subtilis* that can tolerate up to 3 mmol/L Cd and can adsorb Cd efficiently from aqueous solution (Jiang et al., 2009; Wang and Sun, 2013). Pot and field experiments have shown the potential of B38 for reducing Cd mobility in soil and Cd accumulation in some vegetables, especially when B38 is applied together with a biowaste (NovoGro) that can promote the proliferation of B38 (Wang et al., 2012, 2014a, 2014b).

4. Factors influencing the remedial effectiveness of OM

4.1. *OM properties*

Cd content. If the added OM itself has a high Cd content, it is obviously not conducive to the remediation of Cd-contaminated soils and may even aggravate Cd pollution (Wang et al., 2015c). Toxic metals have been detected in many organic additives (Khan et al., 2017). For example, Wang et al. (2015a) analyzed animal manure samples from ten provinces in China and found that the total Cd concentrations ranged from below the limit of detection to 10 mg/kg, with median values of 0.4–1.2 mg/kg, and the Cd concentrations in manures increased significantly from 1990 to 2010. Some rice straws also contain considerable amounts of Cd. Yuan et al. (2019) measured the Cd content of a rice straw to be 0.76 mg/kg. Total Cd in the rice straw produced in heavily contaminated farmland (total soil Cd content 4.57 mg/kg) can be as high as 3.87 mg/kg (Ni et al., 2017). In a soil incubation experiment, Wang et al. (2015c) added rice straw that contained 6.66 mg/kg Cd to paddy soil and found that it significantly increased the total and available soil Cd after 28 days.

pH. If exogenous OM has a pH value higher than that of soil, the addition of exogenous OM may increase soil pH, thereby promoting the adsorption and immobilization of Cd (Hwang and Neculita, 2012); conversely, if the pH of the added OM is lower than that of soil, it is expected that OM addition may decrease soil pH and mobilize Cd. Of course, as mentioned above, the influence of OM on soil pH also comes from the decomposition process after OM is added to soil.

Carbon to nitrogen (C:N) ratio. OM with a smaller C:N ratio can better meet the needs of microorganisms for nitrogen, so it will be preferentially used by microorganisms (Chapin et al., 2011; White, 2006). It has been reported that the addition of plant residues, particularly those with low to moderate C:N ratios, to acid sulfate soil stimulated the increase in soil pH after flooding (Jayalath et al., 2016). Therefore, it is worth testing the hypothesis that the addition of OM with a smaller C:N ratio is more conducive to Cd immobilization in flooded paddy soil. Conversely, if the soil is not fertile and the added OM has a high C:N ratio, microorganisms will compete with plants for nitrogen, hampering crop growth (Chapin et al., 2011; Weil and Brady, 2016).

OM fractions. Some researchers extracted different OM fractions from soil and then added them to Cd-contaminated soils to assess their remedial effects. Soil DOM refers to OM dissolved in the soil solution and is operationally defined as OM that can pass through a 0.45 µm filter membrane (Kalbitz et al., 2000). Soil particulate organic matter (POM) is small plant residues and microbial cell debris (Weil and Brady, 2016) and is operationally defined as OM contained in the soil components with a particle size greater than 0.053 mm (Cambardella and Elliott, 1992). The addition of DOM extracted from soil or sediment to contaminated soil will cause Cd mobilization, because the complexation of DOM with aqueous Cd will hinder Cd adsorption on the soil solid phase (Chen and Chen, 2002; Li et al., 2011). The effects of extracted soil POM on soil Cd mobility and the Cd content in rice grains depends on the amount of added POM and watering conditions (Guo, 2018).

Similarly, if the dissolved fraction is extracted from exogenous organic materials and then added to soil, it will usually lead to Cd mobilization (Chen and Chen, 2002; Li et al., 2007; Wang et al., 1999; Xu and Yuan, 2009). Nevertheless, Li et al. (2007) reported that, unlike the DOM extracted from rice straw, the DOM extracted from pig manure could enhance Cd adsorption by soil, possibly due to its high pH and high molecular weight.

In remedial practices, specific OM components are not usually extracted from soil or organic materials and then added to Cd-contaminated soils. However, sometimes the mobilization of soil Cd after OM application is explained by the DOM contained in or released from the added OM (Cui et al., 2008; Jia et al., 2010; Ni et al., 2017; Shan et al., 2008; Zhou et al., 2018).

4.2. *Soil properties*

Cd content. Soil Cd content will intrinsically influence the remedial effectiveness of OM. For example, Kashem and Singh (2001) investigated the effects of flooding and OM application on Cd solubility in three soils with different Cd contents. Although OM addition decreased the soluble Cd concentration in all three soils compared to the control, the absolute extent of the decrease was greater in the two soils with lower total Cd concentrations.

pH. As mentioned before, if the pH of soil is lower than that of the added OM, OM application would increase soil pH, and *vice versa*. In addition, the growth of microorganisms requires a certain pH range. For example, most sulfate-reducing bacteria need a pH range of 6–8 for optimal growth (Sánchez-Andrea et al., 2013). Under extreme pH conditions, such as in acidic sulfate soils with pH < 4, the addition of OM alone cannot promote sulfate reduction after flooding, and the soil pH must be adjusted at the same time to induce sulfate reduction and further increase in soil pH (Yuan et al., 2015a).

Texture. Soil clay particles can bind OM, thus protecting it from being decomposed by microorganisms (Weil and Brady, 2016). Even for freshly added OM, protection by clay particles may occur (Shi and Marschner, 2013). Therefore, in soil with a high clay content, it may be necessary to add more OM to stimulate microbial reduction (Yuan et al., 2015a), increase soil pH, and promote Cd immobilization. This requires further examination.

5. Impact of straw incorporation on Cd mobility in paddy soil

Due to various influencing factors, OM addition does not always reduce the mobility of Cd in soil (Hamid et al., 2020; Khan et al., 2017). This can be demonstrated by the positive, insignificant, or negative results of straw incorporation in the remediation of Cd-contaminated paddy soils (**Table 3**). Here, we use straw as a prime example to discuss the impact of natural OM on Cd mobility in paddy soil, because “straw is the only organic material available in significant quantities to most rice farmers”, and the management of straw is crucial in agriculture (Dobermann and Fairhurst, 2002). Other organic additives, such as manures and sewage sludge, have also shown inconsistent remedial performance. For the details, the readers can refer to Khan et al. (2017) and Hamid et al. (2020).

Straw incorporation not only returns a large amount of nutrients to soil and improves soil structure but also avoids the air pollution caused by straw burning, so it is commonly implemented in agriculture (Bai et al., 2013; Nie et al., 2019). Usually, rice straw is applied to paddy fields, but sometimes other straws are also used (**Table 3**). As an amendment for contaminated soil, straw is appealing because it is cheap and readily available. One hectare of rice field can produce 2–5 tons of dry straw per season, providing a large amount of OM available on the spot (Verma and Bhagat, 1992).

Both short-term (a few months) and long-term ($>$ 10 years) field experiments show that removing or returning straw from/to the field has insignificant influence on soil total Cd concentrations (Feng et al., 2018; Nie et al., 2019; Tang et al., 2015). However, it has been reported that straw incorporation can promote the transformation of soluble and weakly adsorbed Cd to strongly adsorbed Cd, thereby reducing the bioavailability of Cd in soil and the uptake of Cd by plants. For example, in paddy soil microcosms, the addition of rice straw caused soil pH to rise rapidly after flooding, and the concentration of dissolved Cd thereby dropped from 34 µg/kg to below the detection limit within one week (Yuan et al., 2019).

Table 3 – Effects of straw incorporation on Cd mobility in soil and the Cd content in rice grains.

Straw source	Laboratory/field study	Duration	Effects on soil Cd mobility/availability	Effects on the Cd content in rice grains	Reference
Rice	Soil incubation	120 days	Decrease	NA	Zhang et al. (2001)
Rice	Soil incubation	28 days	Increase	NA	Wang et al. (2015c)
Rice	Soil incubation	13 weeks	Decrease	NA	Yuan et al. (2019)
Maize; kidney bean	Soil incubation	10 weeks	Insignificant or increase	NA	Jia et al. (2010)
Rice	Pot	5 months	NA	Decrease	Zhang et al. (2001)
Rice	Pot	4 months	NA	Decrease	Wang et al. (2015c)
Rice	Pot	NA	Insignificant	Decrease for early rice; insignificant or increase for late rice	Wang et al. (2007)
Wheat	Pot	6 months	Increase	NA	Bai et al. (2013)
Rice; rape	Field	2 years	NA	Insignificant	Duan et al. (2017)
Rice	Field	7 months	Insignificant	Increase for early rice but not late rice	Cai et al. (2018)
Rice	Field	7 months	Insignificant	Increase for late rice but not early rice	Feng et al. (2018)
Rice	Field	90 days	Insignificant or increase	Insignificant	Ni et al. (2017)
Rice	Field	9 years	NA	Increase	Tang et al. (2015)
Rice	Field	10 years	Increase	Insignificant	Nie et al. (2019)
Rice	Field	12 years	Insignificant	Insignificant	Nie et al. (2019)
Rape; wheat	Field	90 days	Decrease	Decrease	Zhang et al. (2016)

NA: not applicable or not available.

In pots with rice plants, Zhang et al. (2001) found that adding rice or wheat straw to soil can promote the transformation of exchangeable Cd to iron and manganese oxide-bound and OM-bound Cd. A similar redistribution of soil Cd after the incorporation of rice straw was observed in a field experiment by Mohamed et al. (2010). Field trials conducted in rice-wheat and rice-rape rotation areas also showed that the addition of wheat or rape straw reduced the concentration of exchangeable Cd in soil and the accumulation of Cd in brown rice (Zhang et al., 2016).

Nevertheless, almost an equal number of studies have reported insignificant or negative effects of straw incorporation on reducing the Cd bioavailability in paddy soil or Cd content in rice plants (**Table 3**). In a pot experiment, rice straw amendment was found to have no significant effects on soil pH, DOC content, or Cd concentrations (Zhou et al., 2018). In another pot experiment, however, the incorporation of wheat straw was observed to decrease soil pH, increase soil DOC and soluble Cd concentrations, hinder rice growth, and enhance Cd uptake by rice plants (Bai et al., 2013). Wang et al. (2015c) reported that the addition of Cd-contaminated rice straw (Cd content 6.66 mg/kg) increased the total and available soil Cd. By contrast, some evidence suggests that the Cd contained in the added straw may not increase the concentration of soil mobile Cd. In a soil incubation experiment, Jia et al. (2010) found that the addition of maize or kidney bean straw (Cd content 4.11 and 3.28 mg/kg) resulted in an increase in NH_4OAc extractable soil Cd by 17%–33% in the early stage of incubation compared to the control. The authors pointed out that this increase in mobile soil Cd was not due to the Cd contained in the added straws, which only accounted for 1.5%–3.8% of the total soil Cd. Instead, the rapid decomposition of the added straws, which released large amounts of organic acids and soluble organic carbon, probably promoted the desorption of soil Cd. Ni et al. (2017) also found in field experiments that the incorporation of rice straw with a Cd content of 3.87 or 0.43 mg/kg had no significant effects on the exchangeable Cd concentration in soil or Cd accumulation in rice grains at the harvest time.

The effect of straw on Cd mobility in soil does not necessarily cause a corresponding change in the Cd content of crops (especially their grains) (**Table 3**). Jia et al. (2010) found in a pot experiment that although straw addition increased the concentration of mobile Cd in paddy soil, the Cd content of the planted cabbage did not increase but even decreased. The reason might be that soil Cd mobilization caused by the straw addition occurred at the early stage when the biomass of cabbage was small and the uptake of Cd was limited. Cai et al. (2018) observed in a field experiment that between treatments with or without straw incorporation, there was no significant difference in the concentration of available soil Cd in the tillering, filling, and mature stages of early rice, but the Cd content in brown rice was higher in the treatment with straw incorporation.

Indeed, field experiments have shown mixed effects of straw incorporation on the Cd content of rice grains (**Table 3**). Short-term field trials conducted in rice-wheat and rice-rape rotation areas in Sichuan province, China showed that the addition of wheat or rape straw reduced the accumulation of Cd in brown rice (Zhang et al., 2016). However, short-term field experiments in Hunan province, China found that straw incorporation significantly increased the Cd content in brown rice of either early or late rice (but not both) (Cai et al., 2018; Feng et al., 2018). Tang et al. (2015) analyzed soil and plant Cd concentrations in paddy fields that had been treated by different tillage and straw management practices for nine years. In the field with conventional plow tillage, compared to straw removal, straw incorporation did not affect total soil Cd concentration, increased the Cd content of rice grains by 20.8%, decreased the Cd content of rice straw by 9.3%, and did not affect the Cd content of rice roots. However, in other long-term (10- and 12-year) field experiments, the Cd concentrations of soil as well as rice straw and grains were not significantly different between fields with or without straw incorporation (Nie et al., 2019).

Taken together, straw incorporation alone does not often reduce Cd mobility in soil or the Cd content in rice grains. Reported factors that may affect the remedial effectiveness of straw incorporation include the type, amount, and Cd content of the added straw as well as the duration of treatment (Jia et al., 2010; Ni et al., 2017; Shan et al., 2008; Zhang et al., 2016). When Cd is mobilized after adding straw to soil, it is generally ascribed to the released DOM that can complex Cd (Bai et al., 2013; Jia et al., 2010; Khan et al., 2017; Ni et al., 2017). The release of organic acid from the added straw may lower soil pH and promote Cd desorption as well (Bai et al., 2013; Jia et al., 2010; Zhang et al., 2001). To achieve better remedial results, further investigation of the mechanisms and factors that regulate the performance of straw incorporation is needed. The combined application of straw and chemical amendments such as lime has been recommended by some researchers (Duan et al., 2017; Ni et al., 2017; Zhang et al., 2016).

6. Conclusions

OM is an important adsorbent for Cd in soil. The addition of natural OM to soil can directly and indirectly affect soil Cd mobility. Added OM can alter soil pH and thus influence Cd adsorption by soil. In paddy soil, Cd mobility is closely linked to iron and sulfur redox reactions. After soil flooding, although the reductive dissolution of iron oxide will cause the release of adsorbed Cd, the formation of secondary iron and sulfide minerals may result in Cd immobilization. Microbial cells, as a part of soil OM, can also adsorb Cd. The addition of OM can stimulate the microbe-mediated reduction of oxidized soil components under anaerobic conditions and mitigate the oxidation of reduced soil species after aeration, thus showing the potential to reduce soil Cd mobility. Considering the huge production of organic materials (such as straw) in agriculture, the use of OM for soil remediation has obvious appeal due to the environmental benefits and low cost. Although there have been successful application cases, the properties of OM amendments and soil can significantly affect the remedial effectiveness of the OM amendments. Importantly, straw incorporation alone does not often decrease Cd mobility in soil or the Cd content in rice grains. The released DOM and organic acid are usually to blame when soil Cd is mobilized after OM incorporation.

When considering natural OM as an amendment for Cd-contaminated paddy soils, careful evaluation and adaptation to local conditions are required. The combined application of OM with chemical amendments may be explored. At the same time, more systematic research is needed to clarify the main factors and mechanisms that influence the remedial effectiveness. Most published laboratory experiments have used soils with unrealistically high Cd concentrations, despite the fact that Cd-contaminated soils (including paddy soils) generally have a Cd content of less than 1 mg/kg (Liu et al., 2016; Wang et al., 2015b; Yuan et al., 2018; Zhao et al., 2015). Soils containing lower concentrations of Cd may require less intensive remedial efforts, and the potential of OM is worthy of further exploration. Much attention has been paid to the impact of OM on Cd behavior in flooded paddy soil. However, Cd mobilization in soil and Cd uptake by rice plants in a paddy field occur mainly during soil drainage and oxidation (Khaokaew et al., 2016; Li et al., 2017). More study is needed to understand the factors and mechanisms, including the potential role of OM, that control the kinetics of Cd release during soil oxidation after drainage.

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Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2020.11.016.

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