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# Speciation, transportation, and pathways of cadmium in soil-rice systems: A review on the environmental implications and remediation approaches for food safety

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ABSTRACT

Cadmium (Cd) contamination in paddy fields is a serious health concern because of its high toxicity and widespread pollution. Recently, much progress has been made in elucidating the mechanisms involved in Cd uptake, transport, and transformation from paddy soils to rice grains, aiming to mitigate the associated health risk; however, these topics have not been critically reviewed to date. Here, we summarized and reviewed the (1) geochemical distribution and speciation of Cd in soil-rice systems, (2) mobilization, uptake, and transport of Cd from soil to rice grains and the associated health risks, (3) pathways and transformation mechanisms of Cd from soil to rice grains, (4) transporters involved in reducing Cd uptake, transport, and accumulation in rice plants, (5) factors governing Cd bioavailability in paddy, and (6) comparison of remediation approaches for mitigating the environmental and health risks of Cd contamination in paddy fields. Briefly, this review presents the state of the art about the fate of Cd in paddy fields and its transport from soil to grains, contributing to a better understanding of the environmental hazards of Cd in rice ecosystems. Challenges and perspectives for controlling Cd risks in rice are thus raised. The summarized findings in this review may help to develop innovative and applicable methods for controlling Cd accumulation in rice grains and sustainably manage Cd-contaminated paddy fields.

## 1. Introduction

Food supply and food safety are major global issues (Lam et al., 2013; Matthews et al., 2021; Santeramo and Lamonaca, 2021). Contamination of agricultural soils by toxic elements and their accumulation in the edible parts of crops have attracted substantial attentions (García et al.,

*Abbreviations:* BW, Body weight; CAL1, Cd accumulation in leaf 1; Eh, Redox potential; EXAFS, Extended X-ray absorption fine structure; HvNramp5, *Hordeum vulgare* natural resistance-associated macrophage protein 5; Nramp, Natural resistance-associated macrophage protein; OsHMA2, *Oryza sativa* heavy metal ATPase 2; OsHMA3, *Oryza sativa* heavy metal ATPase 3; OsIRT1, *Oryza sativa* iron-regulated transporter 1; OsIRT2, *Oryza sativa* iron-regulated transporter 2; OsLCT1, *Oryza sativa* low-affinity cation transporter 1; OsNramp1, *Oryza sativa* natural resistance-associated macrophage protein 1; OsNramp5, *Oryza sativa* natural resistance-associated macrophage protein 5; PCS1, Phytochelatin synthase 1; XANES, X-ray absorption near-edge structure spectroscopy; XRFS, X-ray fluorescence spectroscopy.

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2009; Gupta et al., 2008), and thus efforts to mitigate potential adverse effects on the environment and human health are increasing (Zwolak et al., 2019; Sarwar et al., 2017). Accompanying the fast development of modern agriculture and industrialization, contamination with toxic elements in farmland is becoming more intensive (Hu et al., 2020a; Jiang et al., 2020a). Fertilization, mining activities, and wastewater irrigation are the major contributors to contamination with toxic elements in soils (Kowalska et al., 2018). Due to their non-biodegradability and durability, toxic elements in the soil can significantly disturb normal ecosystem functions, impact natural soil microbial communities, and result in a decline of soil health and fertility (Wang et al., 2007). The uptake and accumulation of toxic elements in crops compromise food security and represent a hazard to human health (Rai et al., 2019).

Cadmium (Cd), as a nonessential element, is potentially assimilated by plants in contaminated areas (Singh and McLaughlin, 1999; Riaz et al., 2021). In rice plants (*Oryza sativa* L.), root uptake is the main pathway for Cd accumulation in grains (Abbas et al., 2019; Yang et al., 2020). Consumption of Cd contaminated rice increases the exposure of the general population to Cd and threatens public health (Jiang et al., 2020a; Li et al., 2021). In many regions of the world, including India, Thailand, China, and Japan, issues with Cd contamination in rice paddies increasingly are recognized. Cd levels in rice grains raise wide food safety concerns (Khan et al., 2017; Hussain et al., 2020; Zhang et al., 2020; Rafiq et al., 2014; Shi et al., 2020). The increased consumption of rice, as well as other cereals, contributes to the enhancement of Cd intake in the United States (Egan et al. 2007; Järup and Åkesson, 2009). Consequently, besides significant efforts to prevent the introduction of new contamination into agricultural ecosystems and the environment at large, ecological solutions and remediation approaches are urgently needed to manage the Cd-contaminated paddy soils and ensure food safety (Peng et al., 2018; Huang et al., 2020a; Zhao & Wang, 2020; Yang et al., 2021a,b; Khan et al., 2021). This requires a deep understanding of the biogeochemical behavior of Cd in paddy soils and the mechanisms involved in the uptake, transport, and transformation of Cd from soil to grains.

Sources of Cd, chemical behavior, accumulation, physiological stress and toxicity towards plants, and remediation methods have been documented in previous studies (e.g., Rizwan et al., 2017; Chen et al., 2019a; Ali et al., 2020a; Kunene et al., 2020; El Rasafi et al., 2020; Khan et al., 2021; Wen, et al., 2021; Wu et al., 2021). Identification of key transporters and their roles in Cd accumulation or detoxification would provide useful information for the development of biological breeding to decrease the Cd levels in rice grains (Yu et al., 2019). The key transporter proteins involved in Cd uptake by rice roots are reported to be similar to those of the essential elements, including iron (Fe), manganese (Mn), and zinc (Zn) (Zhang et al., 2019a). Metal chelators and several organic acids also play important roles in reducing Cd toxicity on essential organelles and macromolecules. Some key detoxification genes and the related mechanisms have been explored (Kaur et al., 2017; Pan et al., 2020). However, it remains largely unknown whether phytohormones are associated with Cd detoxification or tolerance. Selecting rice varieties with low Cd accumulation and irrigation management also may be effective strategies for reducing rice Cd, but these options still need to be further developed (Bao et al., 2019; Li et al., 2017a).

Although there are many reviews about Cd biochemical behavior in soils (e.g., Hu et al., 2016; Shahid et al., 2016; Rizwan et al., 2017; Ma et al., 2021), up to our best knowledge the fate and pathways of Cd in soil-rice systems, particularly the speciation of Cd in paddy soils and its transport and transformation mechanisms from soil to rice grains have not been critically reviewed to date. This review is about (1) the Cd geochemical fractions and speciation as well as the factors governing Cd bioavailability in soil-rice systems, (2) the pathways and transformation mechanisms of Cd from soil to rice grains and the associated human health risks, (3) the transporters involved in Cd accumulation and transport and their vital roles in reducing Cd uptake by rice for food safety, and (4) the effective remediation approaches for mitigating the potential environmental and human health risks of Cd contamination in soil-rice systems. This review therefore can contribute in advancing strategies to mitigate Cd accumulation in rice.

## 2. Health risk of Cd exposure

Cadmium has a half-life of approximately 20–30 years in the human body. Prolonged exposure to elevated environmental levels can cause damage to respiratory organs, kidney function, immune function impairments, metabolic disorders, bone loss, endocrine disorder symptoms, etc. (Table 1) (Alkharashi et al., 2017; Cheng et al., 2019; Fransson et al., 2014; Wu et al., 2020; Wang et al., 2020a). Itai-Itai is a typical disease caused by chronic exposure to elevated Cd concentrations from contamination by mining activities (Aoshima, 2016; Suwazono et al., 2019). It is estimated that about 15% of soil Cd can be transported into rice grains, and the human body receives about 3% Cd from rice grains

**Table 1**
Chronic exposure to Cd contaminated rice and health risk.

| Cd concentration | Location | Results | References |
|---|---|---|---|
| 2.5 μg $kg^{-1}$ BW $week^{-1}$ | Italy | Child group that showed the highest estimated weekly intakes of Cd | (Pastorelli et al., 2018) |
| From 15.5 to 27.1 μg $kg^{-1}$ BW $week^{-1}$ | Xiangtan county, China | Potentially cause an increased non-carcinogenic risk | (Chen et al., 2018) |
| Ranged from 20 to 82 μg $kg^{-1}$ BW $week^{-1}$ | Western Thailand | A significant public health risk to local communities | (Simmons et al., 2005) |
| 15.8 μg $kg^{-1}$ BW $week^{-1}$ | Mae Tao sub-district, Tak, Thailand | Potential development of health impacts in the local residents | (Suwatvitayakorn et al., 2020) |
| Ranged from 430 to 930 μg $kg^{-1}$ BW $day^{-1}$ | 20 provinces in China | Adverse health effects on children (2–14 years) | (Qian et al., 2010) |
| Ranged from 70 to 2030 μg $kg^{-1}$ BW $week^{-1}$ | Xiaogan (Hubei, China) | Significant effects on renal health | (Luo et al., 2017) |
| Ranged from 0.63 and 4.06 μg $kg^{-1}$ BW $day^{-1}$ | Bangladeshi markets | The age groups (2–5 years) and (6–10 years) experienced higher risks than others | (Shahriar et al., 2020) |
| A trend of increasing and then decreasing (278, 313, and 255 μg $week^{-1}$, respectively) in three decades | Shanghai residents from 1988 to 2018 | Increased the Cancer risk and disease burden | (Qing et al., 2020) |
| Average rice-Cd levels were 2,73 μg $kg^{-1}$ BW $week^{-1}$ | 7348 inhabitants of the polluted Jinzu River basin, Toyama, Japan | Increased rice Cd concentration decreased the prognosis for life over a long-term observation in women | (Nogawa et al., 2018) |
| Rice-Cd levels were more than 2.1 μg $kg^{-1}$ BW $week^{-1}$ | Jinzu River basin, Toyama in Japan | Mortality is associated with Cd-contaminated rice in the contaminated area of the Jinzu River basin, Japan | (Nishijo and Nakagawa, 2019) |
| From 1.05 to 5.18 μg $kg^{-1}$ BW $week^{-1}$ | Different WHO regions | It is estimated that dietary cadmium would result in 2064 global deaths and 70,513 disability-adjusted life years | (Zang et al., 2019) |

Note: BW, body weight

(Li et al., 2017b) (Fig. 1). The model can be further developed and serve as a novel and accurate approach to examine the Cd exposure caused by rice consumption. To ensure food safety, many countries and regions have established standards on maximum allowable Cd contents in rice (Hu et al., 2016). For example, the allowable Cd content of rice in Australia, New Zealand, and Iran is 0.1 mg $kg^{-1}$ dry matter. European Union, China, and European Food Safety Authority set 0.2 mg $kg^{-1}$ as the maximum allowable Cd content.

## 3. Cd sources in soil-rice systems

Apart from the natural sources of Cd in ecosystems, anthropogenic release is another important source for soil Cd contamination (Hutton, 1983; Yin et al., 2021). In recent decades, Cd has been released into the environment through various industrial processes, such as burning of coal and waste, smelting of metals, fossil fuel burning, and sewage sludge (Shaheen et al., 2013a; Rigby and Smith, 2020; Cai et al., 2019a; Wang et al., 2021). In many developing countries, Cd-contaminated biosolids including sewage sludge from the industrial wastewater are still a major source of Cd input to soils (Shaheen et al., 2017; Fei et al., 2019). The main sources of Cd input to soil-rice systems include atmospheric deposition, phosphate fertilizers, biosolids, and mine tailings (Li et al., 2020; Wang et al., 2021). In most agricultural soils, disposal of sewage sludges and irrigation are also considered as the key anthropogenic Cd sources (Adriano, 1986; Alloway and Steinnes, 1999; Peris et al., 2008).

According to the estimate of Richardson et al. (2001), the total yearly emitted Cd is almost 41,000 t. Forest fire, vegetation (about 24% of Cd input), airborne soil particles (12% of Cd input), and especially the volcanogenic aerosols (about 62% of Cd input), are the major natural sources of Cd input to the environment (Pan et al., 2010). Fertilizers obtained from phosphate minerals are significant sources of Cd in soil-rice systems, comprising about 56% of the total Cd input from anthropogenic activities. Atmospheric deposition contributed 40% of the Cd content from anthropogenic activities (Liang et al., 2017). In soil-rice systems near some mines and smelters, irrigation is considered a major source of Cd contamination. In addition, acid rain can enhance the uptake of Cd by rice with the alternation of soil pH (Yang et al., 2018).

## 4. Geochemical fractions and speciation of Cd in paddy soils

Soil metals including Cd distribute among different geochemical fractions such as soluble and exchangeable, precipitated with carbonates, occluded in amorphous or crystalline Fe-Mn oxides, associated/complexed with organic matters, and residual fractions (Li et al., 2018a; Liu et al., 2020; Wiggenhauser et al., 2021). The soluble and exchangeable fraction is considered to be bioavailable for plants. The non-residual fractions (i.e., carbonate-, Fe-Mn oxides, and organic matter fractions) would also be bioavailable if the pH and redox potential (Eh) of the soil change (Rinklebe and Shaheen, 2014).

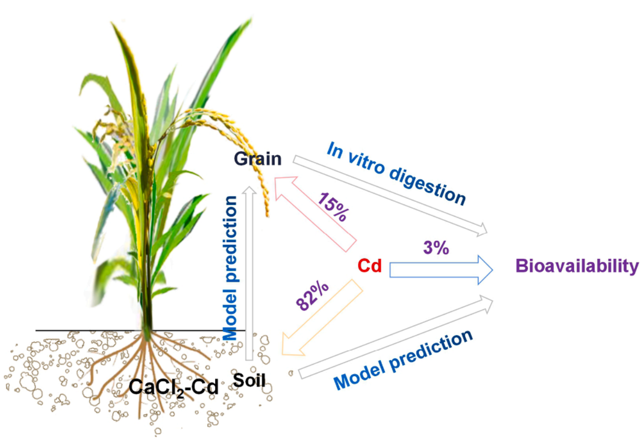


**Fig. 1.** A chain model for prediction of cadmium transfer from contaminated soils to human bodies through rice consumption (Li et al., 2017c).

Generally, Cd is present as a form of complexes with soil colloids (El-Naggar et al., 2018). For rice plants, the bioavailable Cd is represented by $MgCl_2$-Cd and OAc-Cd, while $NH_2OH$-HCl-Cd is the immobilized Cd in soil. In paddy fields, the formation of CdS is associated with the decrease of soil Eh to values lower than −100 mV (Takeno, 2005). In a period of oxidizing conditions, decreasing of sulfide activity leads to an increase of Cd solubility in the soil solution. The distribution of Cd species in paddy soils can be affected by soil pH. A high pH value or increases of phosphate ligands in paddy soils lead to $Cd^{2+}$ hydrolysis and facilitate the precipitation of $Cd(OH)_2$ and $Cd_3(PO_4)_2$. Moreover, in alkaline flooding paddy soil, it has been observed that carbonate-bound Cd is the major species (Mao et al., 2019).

Synchrotron radiation-based technology is of high precision and high resolution and has been widely used in environmental science to analyze the speciation and distribution of trace metals. Thereinto, X-ray absorption near-edge structure spectroscopy (XANES) is typically used for elemental speciation and coordination compounds analyses. Specifically, the XANES spectra can be employed to determine the Cd speciation, atomic structure, and oxidation state in rice plants as well as the information on the bonding of Cd in soil components (Kunene et al., 2020; Siebers et al., 2013). It has been identified that Fe-Mn (oxyhydrogen) oxides fraction acts a prevalent role in controlling the mobilization of Cd upon soil drainage in rice-soil systems, and the decrease of soil pH can cause the dissolution of immobilized Cd from Fe—Mn (oxyhydrogen) oxides complexes (Wang et al., 2019). Yan et al. (2016) reported that different cultivars proposed the change of Cd speciation from Cd—S to Cd—O bonding in rice roots, and the allele-involved Cd accumulation in rice can also be identified with the help of XANES. When the rice is grown in 500 mM $CdCl_2$, the formation of CdS can be identified by XANES analyses (Siripornadulsil and Siripornadulsil, 2013; Hashimoto and Yamaguch, 2013).

XANES is typically used in conjunction with other methods in Cd analysis in contaminated areas. For instance, with the help of soil Eh and mass spectrum analyses in different soil depths it revealed that CdS was accumulated in the subsurface layers of the reduced soil (Su et al., 2016). In rice tissues, the Cd species are identified by XANES to be mainly O-co-ordinated and S-co-ordinated forms in the phloem after microscopic analysis. Additionally, the presence of S-co-ordinated Cd supported that some of Cd is transported to the panicle parts via combining with S-containing compounds (Yamaguchi et al., 2012). As above, XANES can provide sufficient evidence of geochemical fractions and speciation of Cd in contaminated paddy soils. It can be utilized to identify the electronic structure of Cd complexes and the adsorption behaviors of Cd in soil-rice systems. This technology can also be employed to reveal Cd accumulation in rice tissues under different biochemical characteristics of paddy fields.

X-ray fluorescence spectroscopy (XRF) can be employed for the spatial distribution and semi-quantitative analysis of Cd in environmental specimens (Zhang et al., 2013). It was verified by XRF that Cd carbonates were the major species at most flooding periods, while a small number of CdS was found after a long flooding period (Khaokaew et al., 2011). The combined application of micro-XRF and XANES can give more information on the transport, transformation, and accumulation of Cd and its derivatives in plant tissues. Tefera et al. (2020) suggested Cd sequestration in root vacuoles using micro-XRF mapping. A significant amount of Cd is also determined in root hairs, meristematic, dermal, as well as stele tissues. Thus, Cd uptake, transport routes, transformation, and their mechanisms from paddy soils to rice grains would be deeply explored with the application of XANES and micro-XRF in the near future (Kunene et al., 2020).

Moreover, it is important to investigate the speciation and distribution of Cd in rice roots providing a better understanding of the mechanism for Cd accumulation and remediation (Ma et al., 2020). In rice kernels, the information including speciation, atomic structure, and oxidation state determined by extended X-ray absorption fine structure (EXAFS) spectra indicated that Cd clusters were formed from Cd(II)-O with Cd-O bond distance of 2.83 Å (Kunene et al., 2020).

The speciation and distribution of Cd in rice grains critically determine its availability for human exposure (Qiao et al., 2018). It was previously reported presented that Cd-thiolate complexes (66–92%) were the majority in rice grains, and two distribution patterns of Cd in rice, including distribution in the entire grain and the aleurone layer and outer starchy endosperm, were verified by XRF mapping (Gu et al., 2020). The variation of Cd distribution in rice proposes important implications for Cd integration from rice consumption, as well as the management of Cd contaminated paddy fields.

## 5. Factors influencing Cd mobility and bioavailability

Cadmium availability to plants is controlled primarily by its sorption/desorption dynamics, which is mainly affected by soil pH, Eh, and content of soil minerals, e.g., phosphates, metal hydroxides, metal oxides, clays, and organic/inorganic matters (Shaheen et al., 2013b; Li et al., 2018b; Ata-Ul-Karim et al., 2020; Wang et al., 2020b). The mobilization and bioavailability of Cd in paddy soils thereafter significantly affect its uptake and accumulation in rice roots, shoots, and grains.

### 5.1. Soil pH

Cadmium was previously reported to exist in the forms of carbonates, hydroxides, and phosphates, and their solubility and availability increase at low soil pH (El-Naggar et al., 2018). Soil pH is one of the key factors governing the mobility of Cd and thus its absorption by rice (Wen et al., 2020). A negative correlation has always been reported between soil pH and Cd phytoavailability (e.g., Salam et al., 2019; Zhu et al., 2016; Wen et al., 2021). Soil acidity also increases desorption of Cd in soil colloids, facilitating its uptake by roots (Shaheen et al., 2013b). In contrast, at high soil pH conditions, the formation of $Cd(OH)_2$ reduces the mobilization of Cd, thereby resulting in lower Cd accumulation in rice grains (Gong et al., 2021).

### 5.2. Redox potential

Soil Eh plays a key role in controlling Cd solubility (Takeno, 2005; Rinklebe et al., 2016). In the flooded paddy fields, Soil Eh can be highly mediated by oxidized $SO_4^{2-}$, $NO_3^-$, Fe(III), Mn(III/VI), as well as many other soil components which can be utilized by soil microorganisms for respiration (Cui et al., 2020). In the reduction reaction, these substances receive electrons to generate reduced substances, resulting in decreased Eh (Lin et al., 2021). Decreasing soil Eh in flooded paddy soils leads to the reductive dissolution of Fe-Mn oxides, and thus inducing the release of bonded Cd into soil solution and increasing its bioavailability (Shaheen et al., 2016; Wu et al., 2019). On the other hand, reduced Cd can be precipitated in sulfide conditions, which decreases its solubility and bioavailability (Rinklebe et al., 2016). By contrast, in the well-drainage period, Eh may increase which causes dissolution of CdS and release of $Cd^{2+}$ into the soil (Shaheen & Rinklebe, 2017; El-Naggar et al., 2018). The formation of water-soluble $CdSO_4$ may enable rice roots to absorb Cd (Maejima et al., 2007; Han et al., 2018a).

### 5.3. Organic matter

Soil organic matter is one of the key factors governing Cd bioavailability in paddy fields (Sanchez-Camazano et al., 1994; Zeng et al., 2011). As an important adsorbent for Cd in paddy soils, organic matter can be used to immobilize Cd (Yuan et al., 2019), reduce the phytoavailability, and decrease the uptake of Cd in rice. A remarkable decrease of Cd accumulation in rice roots, straws, and grains is previously reported with the presence of organic matters (Ali et al., 2020b). Organic acids, as negative anions, can react with Cd ions and result in immobilization of Cd in paddy soil, thus reducing its phytoavailability (Li et al., 2019a). However, organic matter in paddy soils can also serve as chelates which may enhance Cd phytoavailability (Filipović et al., 2018). It suggests that remediation of Cd-contaminated soils using organic matters should be a double-edged sword, and the application dose of organic matters should be strictly controlled avoiding secondary contamination by toxic elements.

### 5.4. Rice cultivars and other factors

Different rice cultivars perform diverse capacities in Cd uptake and transport (Song et al., 2015; He et al., 2017), and distinct accumulation of Cd among rice cultivars are generally observed in the reproductive stage (Chen et al., 2019b; Zhou et al., 2019). The variations among six rice cultivars in Cd uptake and translocation were investigated and different Cd concentrations in grains among the cultivars were presented (Liu et al., 2007). The solubility of soil Cd can be modulated during Cd uptake due to the root excretion from various cultivars (Filipović et al., 2018; Yang et al., 2021a,b). Low-Cd-accumulating cultivars can potentially decrease Cd phytoavailability in rhizospheric soil (Mei et al., 2020), especially in flooded regimes. Apart from rice cultivars and their root barrier, the irrigation and fertilizer types, the sources and forms of Cd in contaminated regions are important factors influencing the migration and phytoavailability of Cd in rice (Khaokaew et al., 2011). In addition, the competition with common ions, inorganic ligands, metal cations including Zn, as well as essential trace elements would affect Cd accumulation in soil-rice systems (Cai et al., 2019b; Ram et al., 2020).

## 6. Transporters involved in the uptake and transport of Cd in rice

### 6.1. Uptake of Cd in rice

Soil Cd is primarily accumulated in roots (about 79–93%) and then transported upwards. Atmospheric deposition of Cd also leads to a significant increase of Cd concentration in rice grains and leaves, and results in health risk to human beings via rice consumption, although the problem of aerial deposition is usually local-confined to areas with emissions (De Temmerman et al., 2015; Feng et al., 2019). Therefore, both the soil Cd contamination and atmospheric deposition of Cd are critical and should be paid attention to when assessing the ecological and health risks induced by Cd in contaminated rice paddies.

The absorbed Cd may migrate via two channels in opposite directions (Fig. 2). Firstly, the deposited Cd penetrates the rice leaf wax layer and reaches the vascular tissue, after which the phloem carries Cd down to the root and some could be secreted from root surface into soil (Zhou et al., 2020a). Secondly, uptake of soil Cd by root leads to the accumulation of Cd in root vascular tissues through the apoplastic and symplastic pathways. After that, Cd is transported upwards to the rice leaves through the xylem, and deposited in rice grains (Tao et al., 2020). Real-time imaging analysis has revealed the different dynamics of Cd transportation from rice roots to grains under variable soil Cd concentrations (Downie et al., 2015; Ishikawa et al., 2011). Soil-root-grain transport is the key entry point of Cd accumulation in rice. Therefore, identifying the related transporters and clarifying the mechanisms of Cd uptake and transport will open up effective avenues to eliminate Cd contamination in paddy fields (Li et al., 2017c).

### 6.2. Transporters of Cd from soil to rice grains

#### 6.2.1. Soil-to-root transport

The first step of Cd transport in rice is root uptake of Cd from soil. Toxic elements can be hindered into the vascular tissues by Casparian

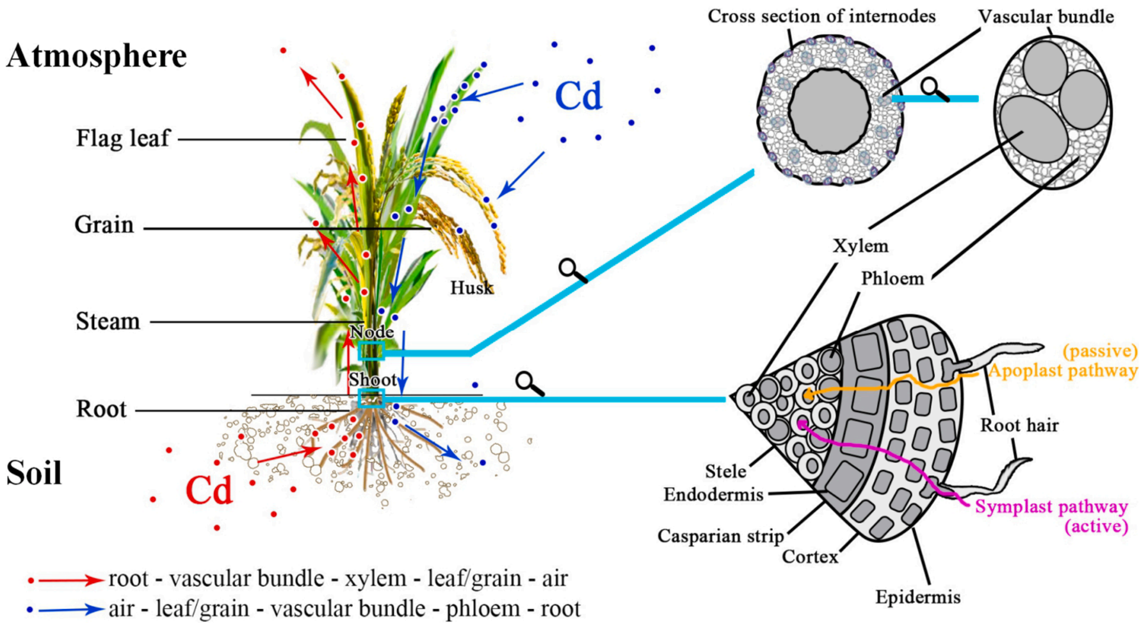


**Fig. 2.** The migration of Cd in the soil-rice system via two channels with opposite direction (Root-to-Air and Air-to-Root).

strips localized in the outer and inner cortex of rice roots. Some specific transporters in the outer cortex (exodermis) and inner cortex (endodermis) of roots are therefore needed in element uptake, including Cd (Fig. 2) (Yu et al., 2020). As a member of the Nramp transporter family (natural resistance-associated macrophage protein), *Oryza sativa* natural resistance-associated macrophage protein 5 (OsNramp5), verified as a major Zn transporter, can carry $Cd^{2+}$ from soil solution into root cells (Ishimaru et al., 2012; Sasaki et al., 2012). The difference in tissue localization of OsNramp5 (Fig. 3) and *Hordeum vulgare* natural resistance-associated macrophage protein 5 (HvNramp5) contributes to the high level of Cd in rice and some other crops (e.g., barley). The higher expression level and transport activity of OsNramp5 in rice than that of other cereal crops also likely contribute to the higher Cd accumulation in rice (Ma et al., 2021). It was reported that knockout of OsNramp5 gene in rice led to an absolute loss of Cd uptake (Tang et al., 2017), and resulted in a significant decrease of Cd accumulation in grains along with hampered Mn uptake under this circumstance (Chang et al., 2020a), indicating that OsNramp5 is an important transporter of Cd in rice.

OsNramp1, *Oryza sativa* iron-regulated transporter1 (OsIRT1), and *Oryza sativa* iron-regulated transporter 2 (OsIRT2) are other three verified transporters that play important roles in Cd uptake in rice (Chang et al., 2020b; Yang et al., 2016). *Oryza sativa* natural resistance-associated macrophage protein 1 (OsNramp1) and *Oryza sativa* iron-

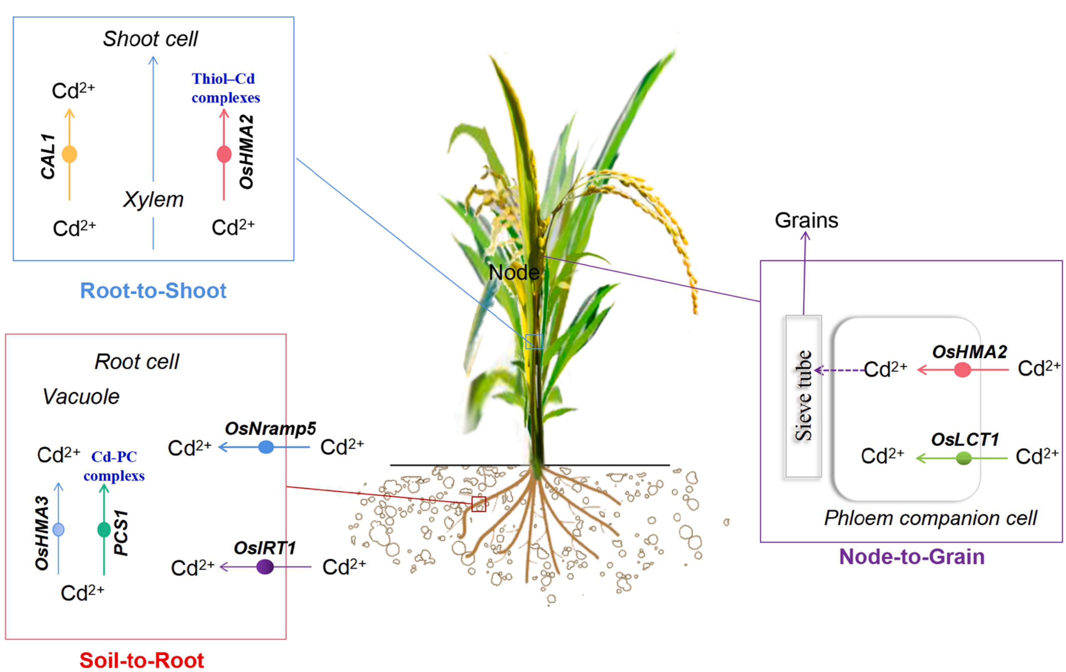


**Fig. 3.** Major transporters involved in Cd uptake and transportation (Soil-to-Root, Root-to-Shoot, Node-to-Grain) and the genes were cited from references (Chang et al., 2020b; Yang et al., 2016; Sasaki et al., 2012; Yan et al., 2016; Ismael et al., 2019; Luo et al., 2018; Wiggenhauser et al., 2021; Yamaji et al., 2017; Liu et al., 2019a; Gao et al., 2016).

regulated transporters (OsIRTs) deficiency had a much weaker influence on Cd uptake than OsNramp5. It is believed that Cd in root cells must be transported toward the xylem vessel with the assistance of the efflux transporters (Jiang et al., 2020b). However, the related efflux transporters in rice have not been completely identified and the involved Cd efflux is still unclear and needs much more studies.

After transport to roots, part of Cd is isolated into the vacuoles (Fig. 3). *Oryza sativa* heavy metal ATPase family 3 (OsHMA3) is critical for Cd transport during the sequestration (Cao et al., 2018). OsHMA3 is mainly expressed in root cells and the influence of Cd stress on it is negligible. However, the transport activity is deficient because of the mutation of one amino acid of OsHMA3. In particular, for high Cd-accumulating cultivars, the loss-of-function allele of OsHMA3 leads to failure of Cd sequestration and causes high root-to-shoot Cd translocation (Yan et al., 2016).

#### 6.2.2. Root-to-shoot translocation

After uptake by roots, Cd penetrates into the xylem and is transported to the shoots with the help of transporters (Fig. 3). *Oryza sativa* heavy metal ATPase family 2 (OsHMA2), localized at the plasma membrane of the pericycle of roots, is pivotal for the root-to-shoot Cd translocation (Ismael et al., 2019). At the vegetative stage, the continuous expression of OsHMA2 in the roots introduces an enhanced influx of Cd. By contrast, Cd contents in straws and grains are decreased by knockout of the OsHMA2 gene (Shao et al., 2018). Recently, it has been confirmed that the Cd accumulation in leaf 1 (CAL1) transporter also mediates the root-to-shoot long-distance transport of Cd via xylem vessels by chelating Cd and facilitating Cd secretion to extracellular spaces, thus decreasing cytosolic Cd concentration (Luo et al., 2018). Interestingly, the CAL1 level is not involved in Cd accumulation in rice grains, and the potential expression and related mechanisms still need further investigation (Sterckeman and Thomine, 2020). Recently, it was reported that the rice without membrane transporter OsHMA3 accumulated Cd in shoots, and the difference in isotope composition between roots and shoots was much smaller than that of rice with membrane transporter OsHMA3, which indicates that the membrane transporter OsHMA3 contributes to the root-to-shoot Cd translocation (Wiggenhauser et al., 2021).

#### 6.2.3. Accumulation of Cd in rice grains via the phloem

Cd accumulation in rice grains is introduced at the reproductive growth stage (Fig. 3). In rice, OsHMA2 and *Oryza sativa* low-affinity cation transporter 1 (OsLCT1) are associated with the intervascular transfer of Cd (Chen et al., 2019c). OsHMA2 is localized at the phloem of enlarged and diffuse vascular bundles at this stage. The Cd content in reproductive organs can be reduced by knockout of OsHMA2 when compared to wild-type rice (Yamaji et al., 2017). Knockdown of OsLCT1 also decreases the content of Cd in phloem and grains, indicating the positive role of OsLCT1 in intervascular transportation of Cd (Liu et al., 2019a). In addition, OsLCT1 might be associated with the efflux of Cd. On the contrary, knockout of the phytochelatin synthase 1 (PCS1) gene can induce Cd accumulation in rice grains partly due to the formation of the phytochelatin-Cd complexes (Gao et al., 2016). However, the exact mechanism remains to be further investigated.

It is promising to develop low-Cd-accumulated rice cultivars based on these principles for the reduction of Cd content in grains. Although transporters associated with Cd uptake and transport in rice are being elucidated, there is still a long way to go before all molecular mechanisms involved in Cd-root-to-shoot-to-grain transport and accumulation are illuminated.

## 7. Management options for mitigating Cd accumulation in rice

Contamination of Cd in paddy fields exposes the public to Cd through consumption of rice. Therefore, it is pivotal to decrease Cd content in rice to mitigate the risk. Reducing the concentration and bioavailability of Cd in paddy soils is one of the key strategies for sustainable agriculture. In recent decades, several remediation approaches have been introduced to treat Cd contamination in paddy soils (Li et al., 2019b; Khan et al., 2021). In this section, some typical remediation approaches were reviewed based on the recent studies, with the aim to shed light on future work to eliminate Cd contamination in rice paddies (Sebastian et al. 2018).

### 7.1. Water management

Management of the water regime in a paddy soil is a relatively cost-effective option allowing to decreasing Cd bioavailability and accumulation in rice. It is well reported that water management had a significant impact on soil Cd lability in acidic soil and the content of Cd in rice. Constant flooding can decrease root Cd uptake during the three growth stages of rice, and the transport and accumulation of Cd in rice are most modulated in the filling stage (Ye et al., 2018; Wan et al., 2018). Flooded conditions decrease Cd phytoavailability, which is possibly attributed to the formation of insoluble CdS (Fulda et al., 2013).

Under the alternately wet and dry irrigation, raw biochar was more conducive in reducing Cd bioavailability compared with the continuously flooded water regime. In paddy soils, Fe-modified biochar treated soils should also be considered because of the increase of uptake and translocation of Cd to brown rice (Wen et al., 2021). Water management can be combined with other amendments to more effectively decrease Cd content in rice (Han et al., 2018b; Rehman et al., 2015; Rizwan et al., 2019). Besides, as reported, the concentrations of arsenic (As) and Cd in rice grain differ due to the different bioavailability of the two elements in soil, as well as the rice cultivars and growing conditions. In paddy soil, the Eh has an opposite effect on the availability of As and Cd, while soil pH exerts more effects on Cd availability than As availability. It remains a great challenge to achieve a simultaneous reduction of both As and Cd accumulation in rice.

Given the above discussion, the soil pH, Eh, and the concentrations of anionic/cation could be modulated by controlling the water regime in paddy, thus decreasing Cd phytoavailability and accumulation in rice. Notably, the safety of S-containing fertilizer application in Cd-contaminated paddy fields should be reassessed in different water regimes.

### 7.2. Fertilization

Fertilization is important for ensuring rice production in paddy fields and can be used as a potential approach in decreasing Cd phytoavailability and uptake by rice. Plant nutrients such as N (nitrogen), P (phosphorus), S (sulfur), Se (selenium), Si (silicon), Fe, and Zn may affect the solubility and bioavailability of Cd to rice.

Nitrogen fertilizers ($NH_4^+/NO_3^-$) improve the concentrations of soluble proteins which increases the fixation of Cd in the rhizosphere, and $NH_4^+$ application decreases Cd phytotoxicity due to high antioxidase activity (Yang et al., 2019). Besides, the application of $NO_3^-$ induces membrane polarization and increases soil pH, which decreases the Cd phytotoxicity (Nogueirol et al., 2018). However, Cd availability in paddy soil may increase due to the decrease of soil pH caused by the excessive addition of $NH_4^+$ (Zhao & Shen, 2018). Application of S fertilizers to paddy fields under flooded conditions can decrease the solubility of Cd as a result of Cd precipitation in the form of CdS under reducing conditions. Sulfate fertilizers also participate in the formation of iron plaque, which reduces the Cd phytoavailability (Cao et al., 2018; Fu et al., 2018). However, the addition of sulfate fertilizers may decrease soil pH and thus increase the mobilization of Cd (Wang et al., 2020c), and sulfide can be oxidized under high Eh and the associated Cd would be released to soil solution in available forms (Shaheen et al., 2016; Shaheen & Rinklebe, 2017). Therefore, the effects of sulfate fertilizers on Cd solubility and bioavailability in paddy soils differ depending on the water regime, redox conditions, and application dose. The wide application of P fertilizers is another important factor influencing Cd phytoavailability in paddy soils (Bairq et al., 2018; Cade-Menun et al., 2017). The necessity of P amendments should be emphasized and carefully evaluated in considering the promoting effects on heavy metals accumulation in grains in initial P-deprived soil (Dang et al., 2016). In addition, besides being a major component of iron plaque, iron-containing fertilizers alleviate oxidative status under Cd stress (Guha et al., 2020). Iron can also compete for the binding site under anaerobic conditions and detoxify Cd (Gao et al., 2016). Se with a high ability to combine with metal ions can counteract the toxicity caused by Cd via decreasing Cd concentration in different rice tissues to recover the root cell viability. Selenate can directly complex with $Cd^{2+}$ in the rhizosphere and promote the formation of iron plaque in the root surface mitigating the accumulation of Cd in rice (Huang et al., 2020b; Wang et al., 2020d).

Zinc can compete with Cd cations for the same transporters, and thus tends to reduce Cd accumulation and related oxidative stress in plants (Huang et al., 2019; Ma et al., 2020a). However, the simultaneous absorption of Zn and Cd is the drawback of Zn fertilizers containing Cd components. Although Si is not an essential element, considerable evidence has suggested that Si is beneficial for rice growth (Rao et al., 2017; Wang et al., 2020d). Several studies have reported that Si contributes to the modulation of tolerance under stress conditions including Cd stress (Hasanuzzaman et al., 2017). The application of Si can detoxify Cd and reduce its accumulation in rice (Hussain et al., 2020). Interestingly, pathways are identified for the reduction of Cd uptake and accumulation in rice by Si application in soil and plants. In paddy soil, application of silicate fertilizers may increase soil pH, mediate the oxidative status, and thus immobilize Cd and decrease its uptake by roots (Dong et al., 2019). For plants, Si application increases photosynthesis through the improvement of mineral nutrients uptake and the fixation of Cd in shoots (Shao et al., 2017). The increase of Si deposition in the endodermis of root cells reduces Cd transport to shoots, through which Cd accumulation in rice grains can be significantly decreased (Zhou et al., 2020b).

Taken together, we can summarize that the combined application of several methods, such as controlling mineral nutrients and fertilizers (N, S, Zn, Se, Si, P), can allow to effectively reduce Cd phytoavailability in rice. However, the application of different fertilizers in rice paddies produces various effects on the accumulation of Cd in rice. To build a reasonable fertilization regime mitigating Cd pollution is still a challenge. It is urgent and significant to reveal the related transporters and transport mechanisms avoiding Cd accumulation in rice impacted by supplement of inorganic and organic fertilizers, which would also help in developing new rice cultivars with low-Cd-accumulation and balanced nutrients (Luo et al., 2020).

### *7.3. Physical-, chemical-, and bio-treatments*

#### *7.3.1. Physical treatments*

Physical removal of the top surface layer in contaminated paddy soil and replacement without contamination is quick and effective *in situ* remediation method. Because of the requirement of unpolluted soil, this approach is not suitable for large areas of soil and the contaminated soil is environmentally disruptive (Hu et al., 2016; Palansooriya et al., 2020). Soil turnover or dilution by *in situ* mixing of the polluted surface soils with the unpolluted subsurface soils may be a cost-effective alternative method (Ahmad et al., 2012), but the subsurface soil usually has low fertility. As an effective soil remediation method, soil dressing is used to prevent access between the rice and the contaminated soil to decrease the uptake of Cd by rice. It should be thick enough to avoid rice roots reaching the covered soil with Cd contamination (Liu et al., 2019b). However, substantial organic fertilizers are necessary to build soil fertility, and irrigation and drainage facilities should be improved.

#### *7.3.2. Chemical remediation via the addition of soil amendments*

Due to the high efficacy of immobilizing reagents, economic feasibility, and applicability, soil amendments including biochar, animal wastes, phosphates, and compost, are widely employed to remediate heavy metal contaminated soils (Palansooriya et al., 2020; Khan et al., 2021). Immobilization of Cd in soils using amendments (e.g., S-/ Si-/ Fe-containing ameliorants) is a very promising approach to reduce the bioavailability of Cd (Palansooriya et al., 2020; Wang et al., 2021). As organic amendments, composts, manures, and biochar are developed to decrease Cd contamination in paddy soils and reduce the accumulation of Cd in grains (Hu et al., 2020b; Hamid et al., 2020). For instance, the adsorption of Cd on biochar and the increased soil pH contributes to the biochar-mediated reduction in Cd uptake and transport in rice (Chen et al., 2019a). The relationship between the long-term application of organic amendments and Cd stability remains unclear and requires further evaluation.

The application of inorganic amendments reduces Cd content in rice mainly by the fixation of soil Cd, with the action mechanism being partly similar to that of organic compounds. For example, lime and sepiolite, magnesium silicate minerals, bentonite, and palygorskite can decrease Cd content in rice by the formation of complexes with added minerals as well as the precipitation of Cd in the form of hydroxides and/or carbonates (Hamid et al., 2019a; Bashir et al., 2020a,b). In addition, the mixtures of several inorganic amendments are more effective in limiting Cd uptake by rice when compared with the separate application (Hamid et al., 2019b). Briefly, the addition of inorganic amendments to paddy fields with Cd contamination is a cost-effective, easily available, and environmentally friendly method.

As shown in Table 2, a comprehensive summary of some relevant studies shows Cd remediation using soil amendments. Both organic and inorganic amendments are introduced and the potential mechanisms or suggestions are presented, providing useful information for Cd remediation in paddy fields. Briefly, abundant, economically feasible, and eco-friendly materials, e.g., processed animal wastes, liming materials, and biochars, are mostly recommended for immobilization of Cd in soil including paddy fields (Antoniadis et al., 2021; Azeem et al., 2021a,b; Yang et al., 2021a,b). Moreover, several nanomaterials have shown potential for environmental remediation due to their high efficiency in removing toxic elements from solution, thus reducing availability to the crop. For example, iron oxide nanoparticles can reduce the Cd concentration in rice tissue (Sebastian et al. 2018). It was also reported that ZnO nanoparticles reduced both As and Cd accumulation in rice grains in co-contaminated paddy fields, supporting new insights for the sustainable application of nanotechnology in the agricultural environment (Ma et al., 2020b). Although the removal of Cd, as well as other toxic elements, by nanoparticles has not been employed on a large scale due to the uncertainty of their safety and economy, it proposes great prospects in the field of soil restoration.

#### *7.3.3. Bio-and phytoremediation*

Phytoextraction, as an effective method to decrease Cd contamination in paddy fields, brings recognized advantages to ecosystems (Antoniadis et al., 2017). Plant species that can absorb Cd to transport and accumulate in shoots can be potentially used to fix and remove soil Cd (He et al., 2020b). Herbaceous species are efficient in Cd uptake and act as key species for phytoremediation. A recent report has revealed that *A. vulgaris*, *S. holostea* and *G. mollugo* can achieve site phytoremediation (Antoniadis et al., 2021). Generally, the application of phytoremediation is more suitable for low and moderately Cd-contaminated paddy fields. Efficient accumulator plants for Cd phytoextraction need to be further designed and developed with the help of molecular bio-techniques. Strategies to efficiently deal with the hyperaccumulator and recover/contain Cd after phytoextraction are still a challenge.

Many soil microorganisms are tolerant to Cd toxicity and can change it to less toxic forms (Siripornadulsil & Siripornadulsil, 2013). Recently, metal-resistant bacteria have been reported to decrease phytoavailability of Cd in soils and its accumulation in plants (Liu et al., 2018; Puga

**Table 2**
The performances of different soil amendments on immobilization of Cd in contaminated soils.

| Amendment type | Amendments | Results or Suggestions | References |
|---|---|---|---|
| Biochar | Rice straw biochar<br>Nut shell biochar<br>Wheat straw biochar<br>Acidified rice husk biochar | Crop types, biochar properties, biochar mixing depth, soil properties, and meteorological factors were involved in Cd immobilization. The applications of biochar also reduced the use of mineral fertilizer. | (Shen et al., 2019; Jing et al., 2020; Qiu et al., 2018; Rehman et al., 2020) |
| Processed animal wastes | Cow manure<br>Egg shell<br>Chicken manure | Chemisorption via processed animal waste in an unexchangeable form was a potential mechanism for reducing Cd mobility and appropriate dosage should be examined to avoid the increase of soil Cd solubility. | (Saengwilai et al., 2017; Huang et al., 2020c; Wan et al., 2020) |
| Compost/ plant residues | Green waste compost<br>Agricultural postharvest waste compost<br>Rice/wheat straw | Varying effects on Cd immobilization resulted from the material maturity levels and composition, as well as soil chemical properties. Moreover, compared with the direct usage of plant residues, the compost can maximize the Cd immobilization. | (Zhang et al., 2019b; Bashir et al., 2020) |
| Biosolids | Sewage sludge | Biosolids acted as an effective sink for decreasing Cd bioavailability in contaminated soils and the effectiveness were mainly associated with soil properties and Cd concentrations. The application of biosolid together with phytoremediation, as well as the conversion of biosolids to biochar, are suggested to maximize the remediation in Cd contaminated soils. | (Chagas et al., 2021; Zuo et al., 2021) |
| Clay minerals | Bentonite<br>Na-Bentonite<br>Sepiolite | Sorption effect has been identified as a possible mechanism for Cd immobilization and the net negative charges of clay minerals is useful for Cd ions immobilization. | (Chen et al., 2020; He et al., 2020a) |
| Liming materials | $CaCO_3$<br>CaO<br>GypsumCa $(OH)_2$ | Liming materials have also been applied as important amendments for minimizing Cd toxicity in soils. Cd carbonates, Cd phosphates, and numerous stable Cd complexes were formed and reduced the Cd bioavailability in soils. | (Meng et al., 2019; Zhai et al., 2020) |
| Metal oxides | Red mud<br>Goethite | Natural and synthesized amphoteric metal oxides and industrial residuals with high surface area and reactive surface sites were widely used as sorbents due to the effective sorption effects. | (Wang et al., 2018; Irshad et al., 2020) |
| Coal fly ash | Coal fly ash | The reduction of Cd caused by coal fly ash primarily through increased soil pH. The efficacy many be effected by soil type, coal fly ash characteristics and metal constituents, and other agro-climatic conditions. | (Munir et al., 2020; Yin et al., 2020) |

et al., 2015). Microbial remediation of heavy metals consists of biosorption, bioaccumulation, biomineralization, and biotransformation (Ayangbenro and Babalola, 2017). Previous studies introduced various bacterial strains that have the potential to deal with Cd-contaminated soils (Liu et al., 2020; Wang et al., 2020e). For instance, soil Cd concentration was highly decreased by the combination of *Serratia* sp. K3 and straw biogas residue via enhancing the extraction of Cd from the soil into plants (Liu et al., 2020). In contrast, *Burkholderia* sp. Y4 inoculation reduced the availability of Cd in the rhizosphere and inhibited its accumulation in rice due to the preferential Cd-sorption by iron–nitrogen coupled cycles (Wang et al., 2020e). Microbial remediation has attracted interest because of its high efficiency, cost-effectiveness, and eco-friendliness (Sebastian & Prasad, 2014; Shan et al., 2019). It can be expected that more efficient and safe engineering microbial approaches will be developed for remediation of Cd-contaminated paddy fields.

### 7.4. Remediation via selecting rice cultivars

Selecting and employing low-Cd-accumulated rice cultivars may be a powerful option to achieve low-Cd rice grains in Cd-contaminated paddy fields (Zhou et al., 2019; Ishikawa et al., 2012). Recently, genetic techniques are being developed for rice breeding to select low Cd rice cultivars with nutritional improvement, nitrogen use efficiency enhancement, as well as herbicide resistance promotion. Point mutations are promising to develop low-Cd-accumulating cultivars (Lan et al., 2019). For instance, the OsLCT1 mutant was reported to produce safe rice grains in lightly Cd-contaminated paddy fields (Liu et al., 2019a). However, practical techniques and knowledge to produce low-Cd rice cultivars remain insufficient and various alleles of Cd transporter genes should be screened using advanced molecular biology techniques for breeding of low-Cd or free-Cd rice.

Based on the above-mentioned remediation trials in Cd contaminated soils, we can summarize that these remediation approaches can be effective under specific conditions with advantages and disadvantages and may leave some effects on the soil after completing the remediation process. The trial efficiency and cost-effectiveness may significantly differ between these practices (Khalid et al., 2017; Palansooriya et al., 2020). More details about the remediation sustainability based on aspects such as socio-economical, environmental, and agricultural are included in Hou et al. (2018). For example, among these methods, the application of soil amendments to immobilize Cd and other toxic metals has been widely employed to remediate contaminated soils, because of its fast and easy application and its commercial viability (Palansooriya et al., 2020). Choosing appropriate and gentle immobilizing agents can deliver cost-effective remediation methods and fulfill the principle of green and sustainable remediation because of their lower life cycle environmental footprints (Hou and Al-Tabbaa, 2014; Palansooriya et al., 2020). However, this method reduces the metal soluble fraction but no decrease of the total content is observed and some amendments may affect negatively the soil properties.

Importantly, among remediation approaches, phytoremediation is one of the nature-based solutions for remediation of Cd and toxic metals contaminated soils and it is the less destructive and drastic process and can achieve the aims of green and sustainable remediation, because it aims to minimize the secondary remediation environmental impact (Hou & Al-Tabbaa, 2014; Antoniadis et al., 2021). Also, phytoremediation is the most viable option for contaminated areas on the scale of hectares, with little ecological disturbance (Antoniadis et al., 2017, 2021). However, the phytoremediation trial has some disadvantages; for example, phytoremediation is slow, prone to trial-and-error cycles, involving local plant species of usually low productivity (Antoniadis et al., 2017, 2021). Antoniadis et al. (2017, 2021) also found that phytoremediation of one $m^3$ may cost about 37 US dollars. Moreover, plants after the completion of the process should be carefully treated, with incineration being a viable option (Antoniadis et al., 2021).

## 8. Conclusions, implications, and challenges

Rice consumption represents a major pathway of human exposure to Cd contamination. Effectively controlling Cd risk in soil-rice requires a thorough understanding of the processes and mechanisms of Cd accumulation in rice plants. Challenges and perspectives for controlling Cd risk in rice are thus put forward. Herein, we reviewed the recent advances in understanding uptake, transport, and transformation mechanisms of Cd from paddy soils to rice grains, the transporters involved in low-Cd-accumulating rice, as well as the mitigation strategies.

The related efflux transporter in rice has not been identified and the involved Cd efflux is to be unraveled which will benefit in low-Cd rice breeding. Therefore, more effective approaches are necessary to identify relevant transporters and provide useful information for breeding in the future. The correlations between mineral nutrients and soil/plant Cd content also must continue to be unraveled to develop low-Cd-accumulation and balanced nutrients cultivars. Moreover, mitigation strategies are in development, and short-term and long-term solutions with cost-effectiveness are needed to cope with the current Cd-contaminated paddy fields and Cd accumulation in rice grains. More importantly, long-term field trials are highly desirable to examine the residual effects of various amendments on different soil types and environmental conditions, and the eco-/health risk and benefits analyses for remediation strategies are also required.

Additionally, assessing the potential of environmentally friendly and low-cost amendments as well as appropriate management of fertilizers and soil conditioners to control the uptake of Cd by rice in paddy soils should lead to practical and applicable recommendations to rice farmers. Moreover, the results will help assess the economic feasibility of the addition of organic and inorganic amendments as well as irrigation water to rice soils, which helps stakeholders and policymakers in demonstrating alternative methods of farming paddy soils and creating new business opportunities for farmers and rice food producers. In short, this review provides a holistic view and would shed light on subsequent studies on developing efficient approaches to mitigate Cd pollution in paddy fields and eliminate Cd accumulation in rice grains to ensure food safety.

Author statement

All the contributors of this work have approved the final version of this manuscript, and declare no financial and personal relationships with other people or organizations that can influence this work.

All the authors of this work also declare that there is no conflict of interest with any other communities or institutes if this manuscript could be accepted.

## CRediT authorship contribution statement

**Zhanming Li:** Investigation. **Yi Liang:** . **Hangwei Hu:** . **Sabry M. Shaheen:** Visualization. **Huan Zhong:** . **Filip M.G. Tack:** Writing – review & editing. **Mengjie Wu:** Writing – review & editing. **Yu-Feng Li:** Writing – review & editing. **Yuxi Gao:** Writing – review & editing. **Jörg Rinklebe:** Supervision, Writing – review & editing. **Jiating Zhao:** Supervision, Writing – review & editing.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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