

# Remediation of Toxic Metals from Paddy Fields

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Toxic metals (TMs) may affect human growth and development, physiological metabolism, etc., and may cause diseases and even death. TMs enter the food chain via organisms located at the bottom of the food chain, and their concentration and toxicity are subsequently amplified as they move further up the food chain. Consuming a certain amount of food contaminated by TMs can threaten an individual's health. Thus, humans (who are at the top of the food chain) face great health risks, as they risk TM exposure principally through food intake. Rice is more important than fish in terms of the risk of metal exposure in the human diet, and arsenic requires particular attention. Grain crops (e.g., rice) that grow on soil/water polluted by TMs not only experience a reduction in yield and quality but also enrich a large amount of TMs. To reduce the threat of TMs to human health, measures must be taken from the source. In particular, uncontaminated soil and water bodies can guarantee the production of healthy food, which is key to human health. Therefore, the research and exploration of the technical methods of heavy metal removal or remediation in rice fields is of great significance to human food safety and health.

Keywords: toxic metals ; health risk ; paddy field ; bioremediation ; rice–fish co-culture system

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## 1. Remediation of Toxic metals (TMs) with Soil Amendments

### 1.1. Metal Soil Amendments

At present, the majority of relevant research is directed toward soil amendments due to their rapid and efficient effects. In particular, soil amendments are a class of compounds containing Ca, Fe, etc. TM adsorption by soil amendments occurs via physical adsorption, surface complexation, and ion exchange <sup>[1]</sup>, which convert TMs to non-bioavailable forms <sup>[2]</sup>. The bioavailability of TMs in soil is the result of the interaction of organic matter, ions, redox conditions, and soil pH <sup>[3]</sup>. Soil pH exerts a great influence on TM content in rice; for example, the optimal pH for the adsorption of Ni (II) and Cu (II) is 6 and 5, respectively <sup>[4]</sup>. pH levels in paddy fields can be increased by applying liming and red mud <sup>[1][5]</sup>. As well as impacting pH levels, red mud also improves the microbial composition in paddy fields and increases the activity of urease, acid phosphatase, and catalase in the soil <sup>[5]</sup>. Fe exhibits high bioavailability and does not exert adverse effects on rice quality and yield <sup>[6]</sup>. Adding Fe to paddy fields can reduce the absorption of TMs by rice and increase the elemental contents of Fe, Cu, Mn in rice grains, and Zn in rice plants <sup>[7]</sup>. Moreover, the application of Fe-containing materials can effectively reduce the concentration of As in soil solutions and rice grains, with zero-valent Fe demonstrated to be particularly powerful. Makino et al. <sup>[8]</sup> attributed this to the formation of arsenic sulfide. Moreover, Yu et al. <sup>[9]</sup> determined a significant positive correlation between As and Fe, suggesting that Fe-containing amendments may have an indirect influence on the fractionation of soil As and biological effects to ease the As for rice. The application of metals or additional complexes can enhance the amount of iron plaque, which is composed of crystallized and amorphous iron oxides, hydroxides, etc. <sup>[10]</sup>, on the root surface <sup>[11]</sup>. This technique can also enhance the interception of TMs by rice roots.

### 1.2. Non-Metallic Soil Amendments

Non-metallic soil amendments are mainly compounds containing silicon (Si), organic matter, etc. Si soil amendments have been the focus of much research due to their ability to actively induce the molecular expression of Cd tolerance in rice leaves. The addition of Si to paddy fields can reduce the As and Cd content in rice, alleviate abiotic stress, and increase rice yields, while also significantly improving the uptake of N, P, and K by rice <sup>[12]</sup>. Immobilized metals are typically bound in soil organic matter components and exist as an organic binding state <sup>[13]</sup>, thus facilitating research on organic soil amendments. Common biochar contains straw, hull, etc. <sup>[14][15]</sup>. Biochar can effectively fix TMs and reduce their bioavailability and mobility in soil <sup>[13]</sup>. Moreover, biochar can directly or indirectly affect indigenous microorganisms by changing the physical and chemical properties and TM content of sediments <sup>[16]</sup>. However, biochar has also been reported to induce oxidative stress in rice <sup>[17]</sup>, and thus any potential negative effects of biochar must be considered in agricultural and environmental applications. Furthermore, the application of non-metallic elements (e.g., Se and S) can alleviate the toxic effects of TMs on rice <sup>[11][18]</sup>.

### 1.3. Nanoscale Soil Amendments

Nanoscale soil amendments have recently been the focus of much research, achieving promising results. For example, Nano-Si has a positive impact on the yield and growth of rice in polymetallic contaminated soil and can reduce TM content in grains [19]. Moreover, CuO nanoparticles have been reported to accelerate the arrival of the rice heading stage, shorten the plant life cycle, and reduce As accumulation in grains [20]. Biochar nanoparticles have a high adsorption affinity for Cd, thus reducing the toxicity of Cd in rice. This is particularly true for biochar nanoparticles prepared under high temperature conditions, manifested as increased biomass, root activity, and chlorophyll content in rice plants [17]. However, the toxicity and outcome of co-existing metals with nanoparticles remain unclear. The negative impact exerted by CuO nanoparticles on plant growth is more significant than that of bulk particles [21]. High ZnO nanoparticle concentrations are able to enhance the content of bioavailable Cd in rhizosphere soil. The addition of ZnO nanoparticles at high concentrations to soil containing low levels of Cd can significantly promote Cd accumulation in rice [22].

### 1.4. Composite Soil Amendments

Multiple TMs are typically present in paddy fields; thus, applying composite soil amendments rather than a single component is required. The Cd bioavailability in the rhizosphere of rice can decrease by 92–100% from the tillering stage to maturity via the application of Ca-Si-rich composite minerals. In addition, Si deposition on the rice root cross-section has been observed to significantly increase following the application of a Ca-Si-rich composite mineral treatment, which consequently enhances the storage of Cd in roots and reduces the translocation of Cd from the root to the shoot [23]. Sulfur and iron-modified biochar amendments can significantly increase the amount of iron plaque on the root surface, facilitating the transition of Cd to binding states (such as Fe-Mn oxide) and reducing Cd concentration in contaminated soil and rice grains [11]. The combined application of biochar and lime reduces Pb availability in soil and Pb accumulation in brown rice at a greater rate compared to the corresponding single applications [24]. Furthermore, integrating ferric oxide and calcium sulfate into a single amendment can effectively reduce the bioavailability of Pb and Cd in soil and the content of Cd, As, and Pb in rice grains [25]. Moreover, Honma et al. [26] demonstrated the ability of prolonged flooding with short-range-order iron hydroxide and rainfed management combined with converter furnace slag to reduce both the Cd and As uptake of rice.

## 2. Bioremediation of TMs

Bioremediation is a low-cost technique that has a limited impact on the environment, and includes phytoremediation, microbial remediation, animal remediation, etc. Crop rotation and intercropping are common TM remediation methods that can effectively ensure the safety and yield of rice and restore metal-contaminated soil [21][27][28], with examples including wheat-rice rotation, oilseed rape-rice rotation, rice-water spinach intercropping, etc. The composition and sources of environmental microbiota play a key role in the health and productivity management of sustainable agriculture. The application of microorganisms to paddy fields contaminated with TMs is a newly developed remediation method. Scholars have identified a reduction in TMs with resistant bacteria in rice grains and have highlighted the potential of bioremediation for contaminated soil. For example, Cd transporters (OsHMA2 and OsNramp5) in rice roots can experience down-regulation following inoculation with *Stenotrophomonas maltophilia*. This may be an internal factor affecting Cd content in rice [29]. Lin et al. [30] determined *Stenotrophomonas acidaminiphila*, *Pseudomonas aeruginosa*, and *Delftia tsuruhatensis* to be Cd tolerant, effectively reducing the enrichment of Cd in rice grains. In particular, *P. aeruginosa* is considered to be a multi-metal-resistant bacterium. Animal remediation technology refers to the absorption, transfer, or degradation of TMs through the food chain of soil animals, and research in this field is relatively limited. The backbone of animal remediation is microbial remediation [31]. For example, earthworms have the ability to alter the structure and permeability of soil, and form the basis of the most commonly used remediation method for TM-contaminated soil.

## 3. Field Management

Water management approaches are easy to operate and are commonly adopted. For example, continuous culture flooding is an effective method for reducing TM content in rice grains [32], yet it has an increased risk of As accumulation [8]. In addition, the aerobic conditions created by the release of water [33] in aerobic treatments can increase Cd concentrations [34]. Flooding in paddy fields may cause sulfide mineral precipitation, significantly reducing trace metal solubility [35], as well as the affinity for metals in the rhizosphere and iron plaque on the root surface [32]. Although the drying-wetting cycles of soil promote the release of metals into the water, Honma et al. [36] determined that intermittent irrigation (3-day flooding and 5-day no-flooding) can simultaneously reduce the accumulation of As and Cd in grains.

Current research on the combined benefits of intermittent and aerobic irrigation demonstrates the ability of intermittent irrigation to reduce the Cd content in grains and increase rice yields [37].

## 4. Planting Methods and Varieties

Furthermore, the cultivation, season, and variety of rice may affect the relationship between rice and TMs during the production process. Deng et al. [38] demonstrated greater Cd and Pb contents in brown rice, straw, and roots via the direct seeding method compared with manual transplanting and seedling throwing. Farooq and Zhu [39] and Yi et al. [40] determined that early and late planting of rice impacted the Cd content in white rice. Differences in rice varieties are attributed to gene differences. Under low and moderate soil Cd pollution, japonica rice cultivars are more suitable than indica rice [41]. Dry season varieties are more tolerant to arsenite or arsenate than rainy season varieties [42]. Studies have also reduced the toxicity of TMs to rice by inserting or removing certain genes via genetic engineering and cross breeding [43][44][45][46]. For example, low Cd accumulation may reduce Cd absorption by inhibiting the bioavailability of Cd in the rhizosphere or by decreasing Cd transport [47]. Thus, the genotype, environment, and their interaction are considered the most significant factors affecting the TM content in rice grains.

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