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Remediation Technology for Combined Pollution of Lead and Cadmium in Farmland Soil

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Abstract At present, the problem of heavy metal pollution in farmland in southern China is serious. Especially, the cadmium and lead are two heavy metal elements with serious pollution and great harm to human body. This paper reviewed some common methods and materials used in the control of cadmium and lead pollution in farmland soil. Then, it discussed the problems in the repair of cadmium and lead pollution in farmland soil. It came up with the future research direction, to provide references for remediation of lead and cadmium pollution in farmland soil.

Key words Farmland, Heavy metals, Combined pollution of lead and cadmium, Remediation technology

1 Introduction

Like water and air, the soil is one of the essential material resources for human survival. However, with the continuous improvement of human productivity and deepening of the urbanization process, the level of heavy metal pollution in farmland soil is constantly deteriorating. According to the survey conducted by the Ministry of Environmental Protection and the Ministry of Land and Resources, among the soil sites surveyed, 16.1% of the polluted sites were affected, of which 19.4% were farmland soil^[1]. The situation of heavy metal pollution in farmland soil is not optimistic. There are many kinds of heavy metal pollutants in the soil, including cadmium, mercury, chromium, lead, arsenic, zinc, copper and nickel^[2]. As the representative of heavy metal pollution, cadmium and lead have strong toxicity and wide pollution range and have received wide attention. Compared with other heavy metal elements, cadmium is the most harmful element to human body. It can cause symptoms such as anemia and neuralgia in the human body, and can cause pathological changes of important organs of the human body. Cadmium can also cause serious damage to the skeletal system and cause abnormal metabolism of calcium. If the cadmium content exceeds the standard in the rice and drinking water, it will cause cadmium poisoning, and the skeletal system will suffer severe damage, forming "bone pain" which once occurred in Japan^[3–5]. Lead is also a heavy metal element that is very harmful to the human body, it can enter the human body along the

blood through various channels and accordingly invade the vital organs and tissues of the human body. Lead can also cause clinical symptoms such as constipation, anemia, vomiting, and nerve injury. Lead damage to children is more serious. Once the amount of lead in children's blood is greater than 60 $\mu\text{g}/100\text{ mL}$, children will have symptoms such as mental retardation and abnormal behavior. Even low concentration of lead may cause permanent damage to the child's nervous system^[6]. There have been extensive studies about the remediation of heavy metal pollution in China, but most of them focus on a single heavy metal element. In this study, we summarized the current remediation methods for heavy metals such as lead and cadmium, in order to provide references for promoting the remediation of multi-metal lead and cadmium pollution in farmland soil.

2 Heavy metal pollution in farmland soil both at home and abroad

According to statistics, the annual emissions of various heavy metals in the world are about 15 000 t of Hg, 3.4 million t of Cu, 5 million t of Pb, 15 million t of Mn, and 1 million t of Ni^[7]. Huang *et al.*^[8] surveyed 1 200 rice soil samples in the United States and found that soil samples polluted with heavy metals such as lead, chromium, cadmium and copper were up to 63%. In Japan, the cadmium contaminated farmland is about 472 000 ha, accounting for 82% of the country's contaminated farmland^[9]. According to statistics of the Ministry of Agriculture, the contaminated farmland of China has been up to 26.67 million ha; in the sewage irrigation area, 64.8% of the farmland has been polluted by heavy metals, the contaminated area exceeds 3.3 million ha, and the annual grain loss is up to 12 million t, and the direct economic loss exceeds 20 billion yuan. Mercury and cadmium pollution is the most serious^[10–12]. According to statistics^[13–17], 32 000 ha of farmland in China has been contaminated by mercury

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ry, and 13 300 ha of farmland has been contaminated by cadmium. At present, 11 provinces and 25 regions are affected, and the situation is serious in the Southwest China, Central China, Pearl River Delta and Yangtze River Delta^[18]. In north China, such as Liaoning Province, the cadmium content exceeded the background value by about 23.02 times; the copper content in Guangdong Province reached 2.92 times the background value; in the southwest China, such as Guangxi, the lead and cadmium content in the soil was also significantly higher than the background value^[19].

Guangxi is an important rice production base in China. It is also a famous "hometown of non-ferrous metals" and China's important non-ferrous metal producing area. The random discharge of mining waste has caused heavy metal pollution in the surrounding and downstream farmland. The current situation of heavy metal pollution of farmland in Guangxi is extremely complex, especially cadmium and lead pollution, so the regional agricultural soil environment and agricultural product safety are very urgent. Some surveys have shown that the problem of excessive levels of heavy metals such as cadmium and lead in paddy fields and rice in Guangxi is extremely serious. Qu Limei *et al.*^[20] surveyed the cadmium pollution of agricultural soil in Xijiang River Basin in Guangxi, and found that 32% of the soil samples in the basin were heavily polluted, and the cadmium exceeding rate of rice grain was up to 23%. Qin Chaoke *et al.*^[21] surveyed the heavy metal elements in southwestern Guangxi and found that the content of heavy metal cadmium and lead in the area was much higher than the background value, and the degree of cadmium pollution was the most serious. Deng Chaobing *et al.*^[22] surveyed the contents of various heavy metal elements in the soil in the karst area of Guangxi. The results showed that the cadmium and lead in the surveyed area were excessive, and some scholars found that the samples of heavy metal cadmium in paddy soil in Guangxi had the excess rate up to 25%. These indicate that the heavy metal cadmium and lead pollution in paddy field of Guangxi have become the main issue of the food security.

3 Current status of remediation of cadmium and lead pollution in soils both at home and abroad

For agricultural products, soil is the medium for their survival. It is extremely important to improve the physical and chemical properties of the soil to reduce or deactivate the toxic substances in the soil. The remediation of heavy metal pollution is to reduce the content of heavy metals in the soil or reduce the activity of heavy metals in the soil by various technical means, so as to restore normal functions of the soil ecosystem and reduce the amount of heavy metal pollutants in the food chain^[23]. According to whether there is change of location of contaminated soil, the remediation can be divided into *in-situ* remediation and *ex-situ* remediation. According to the principles, the remediation can be divided into physical remediation, chemical remediation, biological remediation and joint remediation. In Japan, the treatment of heavy metal cadmium pollution is mainly through Hakka method and leaching, to reduce

and eliminate pollutants. However, this method consumes high energy and needs high remediation costs, so it is not feasible for China. At present, most studies about the remediation methods of cadmium and lead heavy metals are the study of chemical *in-situ* remediation and biological remediation.

3.1 Chemical passivation of heavy metal lead and cadmium pollution in farmland soil In the chemical *in-situ* remediation currently used in China, the chemical passivation technology started earlier. Because of its advantages of short time of remediation and low cost, the chemical passivation technology is a widely used and relatively mature technology. The core of chemical passivation remediation is the addition of various passivating agents into the contaminated soil. It is to reduce the biological reaction of heavy metals through the chemical reaction between the heavy metal pollutants in the soil and passivating agents. Clay minerals, inorganic materials and organic materials are three types of passivating agents that are currently used in the remediation of lead and cadmium contamination.

Through combining zeolite, sepiolite, bentonite, diatomaceous earth and limestone, Zeng Hui *et al.*^[24] carried out the *in-situ* immobilization of heavy metal contaminated soil in the mining area and found that the ratio of diatomite to limestone at 1:2 had the best immobilization effect of heavy metal pollutants in soil; compared with the control group, the content of lead and cadmium in the soil sample extract reduced by 54.3% and 99.0%, respectively. Through simulation experiment, Fu Yuheng *et al.*^[25] studied the passivation effect of potassium phosphate dihydrogen phosphate, dibasic calcium phosphate and ammonium dihydrogen phosphate on the lead and cadmium contaminated soils. The results showed that these three phosphates can significantly reduce the bioavailability of lead and cadmium, and the best effect is calcium dihydrogen phosphate. When $n(\text{P})/n(\text{Pb} + \text{Cd}) = 5:1$, the passivation rate of Pb and Cd can reach 69.81% and 35.18%. The remediation mechanism is that phosphate can form poorly soluble phosphate precipitates and hydroxyphosphorus lead and cadmium minerals with lead and cadmium heavy metal pollutants in the soil. Organic materials include poultry and livestock manure, organic compost, activated sludge, humic acid, and *etc.* Through studying the effects of humic acid on soil cadmium, Wang Jing *et al.*^[26] found that humic acid can significantly reduce the content of soluble cadmium in soil. This is a powerful evidence for humic acid as a soil heavy metal pollution improver. Yang Haizheng *et al.*^[27] studied the effects of compost on the form of cadmium in heavy metal pollution. It was found that when the amount of compost increased, the content of exchangeable cadmium in the soil decreased significantly, indicating that composting could also be used as a passivating agent for heavy metal contaminated soil.

3.2 Biological remediation of heavy metal lead and cadmium pollution in farmland soil Biological remediation is a new remediation technology. Compared with other remediation technologies, the biological remediation technology of soil heavy metal pollution started late and it is not mature enough. However, bio-

logical remediation is characterized by low cost and little impact on the surrounding environment. Thus, in the field of soil remediation, it has been favored by many scholars and has huge potential. Phytoremediation and microbial remediation are the two biological remediation techniques receiving wide attention. The key to phytoremediation technology for heavy metal pollution is to find a suitable heavy metal super-enriched plant. The typical representative is *Pteris vittata* L. found by the team of Chen Tongbin. After two years of *in-situ* remediation, Xie Jingqian *et al.*^[26] found that apart from the enrichment for arsenic, *P. vittata* L. also has strong enrichment ability for lead. In addition, *P. vittata* L. is also highly tolerant to heavy metal lead pollution. Even if the lead content in the soil is high, the biomass of *P. vittata* L. will still not be significantly reduced. Wang Xiaoling^[29] found that addition of EDTA in the soil can strengthen the remediation efficiency of *P. vittata* L. for lead and cadmium pollution.

Microbial remediation technology refers to the technology that reduces the activity of pollutants in the soil by adding microbes to the contaminated soil after cultivating the indigenous or exogenous microorganisms. After the microorganisms are added to the soil, it takes a period of time to properly manage and maintain the amount of added microorganisms above a certain level, in which way it can effectively reduce the bioavailability of heavy metal pollutants. Sulfate-reducing bacteria are a facultative anaerobic bacterium that was originally studied because it will generate H_2S that can cause corrosion in buried iron components. However, it was later found that sulfate-reducing bacteria have a good effect on the passivation of heavy metal pollution of soil. Fan Wenhong *et al.*^[30] found that sulfate-reducing bacteria can convert the exchangeable cadmium in soil into a more stable iron-manganese oxide binding state, thereby reducing the bioavailability of cadmium and remediating the polluted soil. In this process, H_2S produced by sulfate-reducing bacteria will form an insoluble cadmium sulfide precipitate with cadmium in the soil. Through the experiment on the decomposition of lead bismuth by sulfate-reducing bacteria, Xu Yihan *et al.*^[31] found that H_2S produced by sulfate-reducing bacteria will also have chemical reaction with lead in the soil to form insoluble precipitate. Therefore, the sulfate-reducing bacteria can also be used to passivate heavy metal lead contamination in the soil. Apart from the advantages of general biological remediation technologies, microbial remediation technology also has the characteristics of relatively short remediation time. Thus, it has become a hot spot both at home and abroad.

3.3 Agronomic improvement measures In farmland soil, the management of agronomic measures will also affect the absorption of heavy metals such as cadmium and lead by crops, such as irrigation methods, fertilization types and crop characteristics. Irrigation methods may be a major point easily overlooked, but good irrigation methods can really reduce the absorption of heavy metals by crops. Uraguchi and Fujiwara^[32] found that at the late stage of rice heading, if the paddy field was always flooded, the absorption of cadmium by rice could be effectively reduced. Under flooding

conditions, the reducing ability will be enhanced, it will cause free cadmium ions to combine with sulfur to form CdS complex, thus reducing the bioavailability of cadmium in soil. Zhu Haijiang *et al.*^[33] found that the water management of dry farming will lead to an increase in lead content in rice. Improper use of fertilizers also increases the activity of heavy metal contaminants in the soil, resulting in increased levels of heavy metals in the crops. For example, the use of urea results in a significant increase in the accumulation of heavy metals such as lead, cadmium and copper in contaminated soils such as *Lactuca sativa* and *Lolium perenne* L. Atafar *et al.*^[34] found that long-term use of mineral fertilizers can significantly increase the content of cadmium and lead in soil. Jalloh *et al.*^[35] found that the effects of different types of nitrogen fertilizers on the accumulation of heavy metal cadmium vary widely. Therefore, in the actual production, it is required to implement rational fertilization, to avoid excessive use of heavy metals such as cadmium, lead and promote heavy metal cadmium, lead absorption of fertilizers.

Because of the significant differences in the absorption and accumulation of heavy metals between different crops, screening low-accumulation crop varieties is also an effective means to reduce the risk of heavy metal pollution. Rice varieties with low accumulation of cadmium and lead have been screened out. In the future, there will be more varieties with low accumulation of heavy metals screened out to provide a strong guarantee for human health.

4 Existing problems

Now, the remediation and development of farmland soils have made great progress and achieved many results. However, there are still many problems. In terms of heavy metal pollution passivation, there are still relatively few economical and efficient passivating agents that can be used on a large scale and in a wide range. Some passivating agents perform well in small laboratory experiments, but there are always some factors influencing their large-scale production. For example, the iron oxide discovered by the Li Fangbai Research Institute of the Guangzhou Institute of Soil Science has a strong adsorption effect on heavy metal cadmium and lead, and a pilot test was carried out in the paddy field in Hechi and Yizhou of Guangxi. However, the current process of obtaining vegetation for iron oxides is very complicated, so its application in large-scale production management is still very difficult.

Biological remediation has the characteristics of low cost, good remediation effect and no secondary pollution, and has become a hot spot of scholars both at home and abroad. However, because it started late, the technology is not so mature, so there are many problems to be solved. In terms of phytoremediation, the biggest problem is that the long time of remediation. It takes a year or even a few years to use phytoremediation technology. Although China has a vast territory, its population is large, and the farmland area has been greatly reduced by urbanization, it is impossible to use phytoremediation technology on a large scale. Most

of the microbial remediation technologies still remain at the laboratory stage. Although they are performing well at present, once they are put into practical application, the effect should be greatly reduced. Only bioremediation technology can't handle high concentrations of heavy metal contaminated soil. High concentrations of heavy metal pollution can significantly reduce the biomass of the organisms used and even directly kill the organisms used. Therefore, the large-scale application of biological remediation technology has a long way to go.

5 Prospects

At present, China's farmland is seriously polluted by heavy metals, and the situation is complicated. In the case of heavy metal contaminated farmland, a variety of heavy metal pollutants coexists. However, many studies are currently directed at only one type of pollutant, and the results obtained may have deviation in practical application. In the future, research on the remediation technology of contaminated farmland should be strengthened for the simultaneous presence of multiple heavy metal pollutants.

At this stage, the heavy metal pollution situation is complex, so it is impossible to solve such complicated pollution situations with a single remediation technology or material. Therefore, in recent years, there have been more studies on joint processing technology, and joint remediation technology will become the future development trend. When the soil is highly contaminated by many types of high concentration of pollutants, the passivating agents used only by chemical passivation technology are too toxic and too irritating to the environment; by contrast, only using the biological remediation, few living organisms can survive and it takes too long time. If the chemical passivation and biological remediation is combined, it not only shortens the treatment time, but also reduces the impact on the environment, and reduces the risk of secondary pollution. In addition to the combination of remediation technologies, remediation technologies materials can also be combined. According to the characteristics of the remediation materials such as clay minerals, inorganic materials, organic materials and the chemical reactions that occur between them, a certain proportion is combined to form a new remediation material. This can also greatly improve the remediation effect of heavy metal pollutants in the soil. There have been extensive studies about the combination of those remediation materials. Now, based on the existing studies, it is necessary to find a more suitable and accurate combination ratio, further optimize the use conditions of the remediation materials, and improve the remediation efficiency.

China has a vast territory, and the pollution level faced by various regions are not the same. Due to serious pollution, the environmental protection of soil cadmium pollution was carried out earlier in Huanjiang County, Hechi City of Guangxi. In the local area, the method of phytoremediation and even chemical passivation remediation has been tested and applied, but the overall cost is quite high, and the phytoremediation by-products are difficult to treat, and the farmers' acceptance is low. In many areas of

Guangxi, it is planting crops directly in polluted farmland. Therefore, it is the development direction to find out suitable remediation technology for local areas, develop new remediation products using local original resources, and reduce the remediation costs.

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