Combined application of low-accumulation crops and amendments on remediation of Cd-contaminated soils

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Abstract. A total of 51 studies were screened and obtained from databases including China National Knowledge Infrastructure (CNKI) and Web of Science (Thomson Reuters, NY, USA), which focused on effects of the combination of low Cd accumulating crops and amendment on Cd-contaminated soils. Both the available Cd content in soil and the Cd target rate of crop edible parts were investigated from three aspects, namely, the types of soil Cd contamination risk, soil remediation method, and types of amendment. All of the statistical analysis and figures were conducted and prepared using IBM SPSS Statistics 22.0 and Origin 2018 software, to obtain the changes of four indicators including the drop of available Cd content in soil, the drop of Cd content of crop edible parts, growth rate of crop yield and target rate of crop edible parts. Results showed that, the Cd content of agricultural products from low-Cd risk soil can meet the standard of ‘GB 2762-2017 national standards for food safety, limits of contaminants in food’ by planting low Cd accumulating crops alone. For medium-Cd risk soil, the remediation effect (96%) of combined treatment with low-accumulation crops and amendments was obviously better than that of low-accumulation crops alone (18%), and the organic-inorganic compound amendments showed the best comprehensive remediation effectiveness. The Cd target rate of crop edible part from high risk soil can be improved by combination of low accumulating crops and amendment (~17%), however, the land use risk is too high that its use needs to be strictly controlled. This study can provide theoretical basis for the safe utilization and engineering remediation of Cd contaminated soil.

1. Introduction
In the past 20 years, heavy metal pollution continued to rise in China's main grain-producing areas, and the point exceeding rate increased from 7.16% to 21.49%, among which the cadmium (Cd) pollution is the most significant (increased by 16.07%)[1]. Soil-bound heavy metals, especially Cd in agricultural soils can easily transfer to vegetables and grains, which pose a risk to human beings and animals who consume the agricultural products that exceed usable standards[2,3,4]. To promote the sustainable development of agroecosystem and human society, it is of great significance to reduce the availability of Cd in polluted soil, and to develop safe utilization plans for soil with different Cd levels[5,6,7].
Breeding crops varieties with low accumulation of heavy metals is considered to be an effective way to reduce the introduction of heavy metals into human diet[8], which was firstly studied by researchers as early as the 1970s, since the Codex Alimentarius Commission (CAC) of the World Health Organization (FAO/WHO) of the Food and Agriculture Organization of the United Nations (FAO/WHO) discussed world standards for cadmium in various foods in 1972. In recent years, scholars at home and abroad mainly focus on inter-specific and intra-specific differences in Cd absorption and accumulation ability of crops, as well as screening and studying Cd low accumulation crop species or varieties, e.g. Ueno et al. studied 146 rice samples and found that there was a 13-fold difference between the highest Cd accumulation and the lowest Cd accumulation in rice abodes[9]. Scholars further studied the mechanism of Cd uptake by low-accumulation crops and the application of these crops in Cd contaminated soil remediation[10,11]. However, it is found that low-accumulation crops are not always effective in practical application, and in-situ chemical passivation or other agronomic techniques is also needed to reduce the migration and accumulation of heavy metals to the ground part of crop plants[12]. Nevertheless, there are great differences and uncertainties in the effects of their combination.

At present, remediation of farmland polluted by heavy metals has began in China, multiple pilot restoration projects across the country are under way. Research results of low-accumulation crops and in-situ passivation are urgently needed to guide the remediation of soils with medium and heavy Cd contamination. Due to the wide variety of amendments, the role of different types of amendments on low-accumulation crops in soil remediation of medium and heavy Cd pollution farmland is still unclear. In this paper, research results at home and abroad on the combined application of low-accumulation crops and chemical amendment in recent years were reviewed, we discussed the changes of available Cd content in soil, the Cd target rate of crop edible parts, and crop yield in soils contaminated with different degrees of Cd under the conditions of planting low-accumulation crops alone and applying different types of amendments in combination with low-accumulation crops, which will provides reference for the realization of "production while remediation" of soil contaminated by Cd in different degrees.

2. Materials and methods

2.1. Literature retrieval
This paper reviews a range of key studies concerning effects of the combination of low-accumulation crops and amendment on Cd-contaminated soils. Here, 51 papers published between September 2008 and July 2019 are reviewed. China Knowledge Resource Integrated (CNKI) and Web of Science are selected as the search database. First of all, the key words "cadmium" or "Cd" and the abstract "low accumulation" or "low concentration" are used for advanced retrieval in CNKI. Secondly, to search the Web of Science database under the theme of "low accumulation cultivars amendment", and to refine the results using "Cd" and "cadmium". Thirdly, from the final retrieval results, the literatures on the simultaneous use of amendments (or conditioners) and low-accumulation crops for soil remediation are screened out. Finally, sorting out one by one, eliminating duplicates, irrelevant titles, multiple drafts, newsletters, conference reports, etc., and 51 papers are determined and discussed, in full.

2.2. Data extraction and processing
(1) The information was collected as follows: amendment type, low-accumulation crop type, cadmium content in soil, available cadmium content in soil, cadmium content in edible part of crops, crop yield, etc. All screening data were set as control group (CK) and experimental group (T). The control group only planted low-accumulation crops and did not apply any amendment, while the experimental group both planted low-accumulation crops and applied amendment. The types of low-accumulation crops and amendment combined with application collected in this paper are shown in Table 1.
Table 1 Types of low Cd accumulating crops and the relevant amendments in Cd contaminated soil

| Soil type          | Low accumulation crop type | Crop type                        | amendment                                           |
|--------------------|-----------------------------|----------------------------------|----------------------------------------------------|
| Low-Cd risk soil   | Paddy                       | Jiazao 17, Xiangwanxian 13,     | Lime, biochar + lime, bioorganic fertilizer + lime,   |
|                    |                             | Liangyou 6206                   | porous ceramic nanomaterials, phosphate rock powder | |
|                    | Sweet potato                | Zhejiang purple 3               | Lime, biochar, soil conditioner, seaweed organic    |
|                    |                             | Late japonica rice varieties Jia| fertilizer, sepiolite, humic acid, sepiolite +     |
|                    |                             | 33, Xiang Early Indica 24,      | humic acid                                         |
|                    |                             | Xiushui 09, Xiushui 03, Xiang   | Sepiolite, bentonite, chicken manure, sepiolite +   |
|                    | Paddy                       | late Indica 12, Fengyou 9       | chicken manure                                     |
| Medium-Cd risk soil| Oilseed rape                | Hua-chun 2, Hua-Lu 2, Keiko     | Biochar, humic acid                                |
|                    |                             | Kawada                          |                                                   |
|                    | Chinese cabbage             | Hong Kong Mid-Season Black      | Biochar, humic acid                                |
|                    |                             | Leaf Cabbage (XGSJ)             |                                                   |
|                    | Corn                        | Marble 5, Huidan 4, Zhongyu 5   | Organic carbon source, rapeseed cake, pig manure,   |
|                    |                             |                                 | sulfur fertilizer, biochar, sepiolite, palynelia    |
|                    | Paddy                       | Late japonica rice varieties Jia| stone                                              |
|                    |                             | 33, Jiaxing -33, H You 518,    |                                                    |
|                    |                             | Zhejiang you 1                  |                                                    |
|                    | Chinese cabbage             | Love pepper black leaf 333      | Biochar, humic acid                                |
| High-Cd risk soil  | Corn                        | Rompin 206                      | Potassium dihydrogen phosphate, chicken manure     |
|                    | Wheat                       | Wheat 1279-9                    | Bamboo Charcoal                                    |
|                    | Low-accumulation vegetable  | Qingqing 91-5C, oily green 80   |                                                    |
|                    |                             | days of Chinese cabbage, K2     |                                                    |
|                    |                             | green stalk large leaf water    |                                                    |
|                    |                             | cabbage                         |                                                    |

2.2.1 Calculation of indicators

From the perspective of agricultural production and agricultural product safety, the content of heavy metals in edible parts of crops can directly reflect the biological availability of heavy metals in farmland soil and farmland remediation effect [13]. Considering the ultimate goal of Cd contaminated soil restoration is to reduce soil Cd activity, ensure the safety of agricultural production, this paper select Cd content of soil effective state, Cd content in edible parts of crops, crop yield and Cd compliance rate in crop edible parts of the four index to evaluate the combined reconditioning effect of low-accumulation crops and amendment. In the analysis, the two cases of planting low-accumulation crops alone and applying low-accumulation crops together with amendment are mainly compared. The indicators are as follows:

2.2.1.1 The decrease amplitude of available Cd content in soil \( (K_s) \) is calculated as follows:

\[
K_s = \frac{C_{CKs} - C_{Ts}}{C_{CKs}} \times 100\%
\]  (1)

Where \( C_{CKs} \) is the available Cd content in the control group (mg·kg\(^{-1}\)); \( C_{Ts} \) is the available Cd content in the experimental group (mg·kg\(^{-1}\)).

2.2.1.2 The reduction of Cd content in edible parts of crops \( (K_e) \) is calculated as follows:

\[
K_e = \frac{C_{CKe} - C_{Te}}{C_{CKe}} \times 100\%
\]  (2)

Where \( C_{CKe} \) is the content of Cd in the edible part of the control group (mg·kg\(^{-1}\)); \( C_{Te} \) is the Cd content of edible parts of crops in the experimental group (mg·kg\(^{-1}\)).
2.2.1.3. The growth rate of crop yield \((K_y)\) is calculated as follows:

\[
K_y = \frac{Y_T - Y_{CK}}{Y_{CK}} \times 100\% \quad (3)
\]

Where \(Y_{CK}\) is the yield of agricultural products in the control group (kg\( \cdot \)(m\(^2\))-1 or kg\( \cdot \)(667m\(^2\))-1); \(Y_T\) is the yield of agricultural products in the experimental group (kg\( \cdot \)(m\(^2\))-1 or kg\( \cdot \)(667m\(^2\))-1).

2.2.1.4. The reaching rate of Cd content in edible parts of crops \((R)\) is calculated as follows:

\[
R = \frac{n}{N} \times 100\% \quad (4)
\]

Where \(n\) is the number of groups in which the Cd content of edible parts of crops is lower than the standard limit. \(N\) is the total number of groups. The reaching value here refers to the limit value of pollutants in food stipulated in 《GB 2762-2017 National Standard for Food Safety Limits of Pollutants in Food》.

3. Results

3.1. Analysis of soil remediation effects with different Cd risk levels

The research scope of soil Cd concentration is relatively broad. In order to facilitate the classification and sorting of existing literatures, the 《Soil Environmental Quality Standard for The Control of Soil Pollution Risk in Agricultural Land (Trial) (GB15618-2018)》 is referred to. (1) When the Cd content in soil is equal to or lower than the screening value of soil pollution risk in agricultural land, it is denoted as low-Cd risk soil. (2) When the Cd content in the soil is higher than the screening value of soil pollution risk in agricultural land and equal to or lower than the control value of soil risk in agricultural land, it is denoted as medium-Cd risk soil. (3) When the Cd content in soil is higher than the risk control value of agricultural land soil, it is denoted as high-Cd risk soil [14]. Since papers on low-Cd risk soils is less in quantity, this paper focuses on the analysis of the remediation effect of medium- and high-Cd risk soils.

In general, for different types of Cd risk soils, the reduction of available Cd content in soil and Cd content in edible parts of crops after the combined application of low-accumulation crops and amendment was significantly greater than that of low-accumulation crops planted alone. At the same time, the overall effect of medium-Cd risk soil after remediation is better than that of high-Cd risk soil. Firstly, the reduction of Cd available content in medium-Cd risk soil was slightly higher than that in high-Cd risk soil, with median of 42.12% and 35.65%, respectively (Fig.1(a)). Secondly, in medium-Cd risk soil and high-Cd risk soil, the average decline range of Cd content of edible parts of crops was 53.56% and 43.02%, respectively, indicating that the absorption of Cd by low accumulation crops was significantly reduced after the application of amendment in medium-Cd risk soil (Fig.1(b)). Thirdly, the crop yield growth of medium-Cd risk soils was significantly higher than that of high-Cd risk soils after the combined application of low-accumulation crops and amendment. With the exception of some amendments, the application of amendments in medium-Cd risk soils could increase crop yield to different degrees, with the maximum increase reaching 130.9%. However, in high-Cd risk soils, crop yield growth was low or even reduced, as shown in (Fig.1(c)).
In medium-Cd risk soil, additional application of amendment can effectively reduce the available Cd content in soil, thus reducing the Cd content in edible parts of crops. Literature data showed that the Cd target rate of crop edible part in the control group is only 18%, while that in the experimental group was as high as 96% (Fig. 2). This indicates that for medium-Cd risk soil, planting low-accumulation crops alone is difficult to meet the requirements of agricultural safety production. The combined application of low-accumulation crop and amendment can effectively reduce the Cd content of crop edible parts, so as to achieve the purpose of agricultural safety risk control.

For high-Cd risk soils, the Cd target rate of crop edible parts in the control group was 0%, while that in the experimental group was only 17%, indicating that in most cases, agricultural products produced from high-Cd risk soils after joint remediation have food safety risks.
3.2. The influence of amendment type on the remediation effect of soil with Cd risk

First of all, both inorganic and organic amendment can reduce the available Cd content in soil. From the data distribution interval, the combined application of inorganic amendment and low-accumulation crops can reduce the available Cd content in soil by 4.2%–90.57%, the highest value is significantly higher than that of organic amendments (53.95%), but the median and average value is only slightly higher than that of organic amendments, indicating that the probability of occurrence of high value under the action of inorganic amendment is not great. The reduction range of available Cd content is maintained at 35%–86.79% under the application of organic-inorganic compound amendment, the highest value is lower than that of single inorganic or organic amendment, but the lowest value is higher than that of single inorganic or organic amendment, and the stability and mean value of data samples are better than that of single inorganic or organic amendment (Fig.3(a)). In addition, there is no significant difference between inorganic and organic amendments in the reduction of Cd content in crop edible part, which fluctuate between 12% and 87%, while the reduction rate of Cd content in crop edible part by application of organic-inorganic compound amendment is relatively stable, maintaining between 35.26% and 70% (Fig.3(b)). It can be seen that all the combined application of low-accumulation crops and inorganic amendments, organic amendments or organic-inorganic compound amendments can reduce the available Cd content in soil and the Cd content in crop edible part, compared with the simple planting of low-accumulation crop. What's more, from the effect of amendments on crop yield (Fig.3(c)), it can be seen that inorganic amendments are not conducive to crop yield increase, and some amendments may even lead to crop yield reduction. Organic and organic-inorganic compound amendment can significantly increase crop yield, and organic-inorganic compound amendment has the best yield increase effect, with a median yield increase of 83.32%.

The target rate of heavy metals in crop edible part is the main standard for screening the safe utilization technology of polluted farmland [15]. According to the statistical results (Fig.3(d)), when low-accumulation crop are planted alone, the average Cd target rate of crop edible part is lower than 40%. After the addition of amendment, the average Cd target rate in crop edible part is greatly increased to more than 90%. Among them, the Cd content in crop edible part is 100% up to the standard after the treatment of inorganic or organic-inorganic compound amendments, indicating that low accumulation crop combined amendment has good application prospect and promotion value in medium-Cd risk soil remediation.
4. Discussion

4.1. Remediation of low- and high- Cd risk soils

There are few literatures on the use of low-accumulation crops in the remediation of low-risk Cd contaminated soil. However, consistent results show that planting low-accumulation crops alone can make the content of Cd in edible parts of crops lower than the standard limit, and the application of amendment can further reduce the Cd content in edible parts of crops. This indicates that for soils with low Cd risk, the application of low-accumulation crops alone or in combination with amendment can achieve the purpose of risk control of heavy metals in soil. Besides, Most amendments have increased crop yields, except for lime, which may reduce crop yields [16]. Therefore, for the remediation of low-Cd risk farmland, some low-accumulation crops can be selected for alternative planting, and amendment can be added in consideration of crop type and soil texture.

For the remediation of soil with high-Cd risk, the content of available Cd in soil and the content of Cd in edible parts of crops can be effectively reduced by combined application of low-accumulation crops and amendment. However, the Cd content of agricultural products is far beyond the standard limit, posing serious agricultural safety risks. Besides, crop loss was also observed in soils with high-Cd risk, which is the result of excessive concentration of Cd damaging the plant protein, DNA and other structures [17]. Therefore, the use of high-Cd risk soil should be strictly controlled, and possible utilization way include replacing crops with non-edible crops, or returning farmland to forest or grassland [18]. In addition, the content of Cd in soil can also be reduced through plant extraction or enhanced extraction [19], to further realize safe production of high-Cd risk soils.

Figure 3. the drop of available Cd content in soil (a), the drop of Cd content in edible parts of crops (b) and growth rate of crop yield (c), and average Cd target rate of crop edible parts (d), after combined treatment of different types of amendment and low Cd accumulating crops
4.2. Remediation of medium-Cd risk soils

The remediation effect of medium-Cd risk soils was influenced by both low-accumulation crops and amendments. Considering that the variety of crops suitable for cultivation are different in different types and properties of soil, and that Cd uptake and accumulation by crops not only depends on genotype, but also is affected by soil environmental factors, such as Cd bioavailability in soil, soil pH value and organic matter, soil cation exchange capacity, and ion antagonism [20, 21, 22]. The passivation materials can not only react with heavy metals to reduce their bioavailability, but also change or affect the above factors in soil to a certain extent [23, 24], thus affecting the Cd uptake by low-accumulation crops.

According to the statistical analysis from Leon et al. [25], the highest passivation efficiency of inorganic amendment to Cd is higher than that of organic amendment. Similar results were observed from combined treatment of amendments and low-accumulation crops, that is, the highest decrease of Cd content in soil occurred in the inorganic amendment group, which indicates that the influence of amendment type on Cd availability is relatively consistent under the condition of single application of amendment or the combination of amendment and low-accumulation crops. However, although inorganic amendments such as lime can significantly reduce the availability of heavy metals in soil in the short term, their effect on heavy metals in soil is not lasting and requires continuous application. Meanwhile, lime reacts with CO₂ to form calcium carbonate precipitation, which leads to soil hardening and inhibition of microbial activities, and ultimately leads to crop failure (Fig.3(c) [26]. In general, the organic-inorganic compound amendment combines the advantages of both types of amendments and has a good effect on the available Cd in the soil.

The decrease extent of Cd content in edible parts of crops is not consistent with that of available Cd content in soil. Although the passivation effect of organic amendments on soil Cd is significantly lower than that of inorganic amendments, the decrease extent of Cd content in edible parts of crops is similar to that of inorganic amendments. When organic amendment react with Cd in soil, organic material provides a large number of surface functional groups (e.g. carboxyl, hydroxyl, carbonyl) for the adsorption or complexation of Cd in soil, besides, it affects soil adsorption and desorption of Cd by changing soil aggregate structure, soil pH value, CEC and other properties [27]. In conclusion, under the application of organic amendment, the uptake of soil Cd by crops is influenced by the content of available Cd in the soil, soil properties, as well as the interaction between organic matter and the root microenvironment[28], while the specific mechanism and the passivation contribution rate from each process are still unclear.

When the organic-inorganic compound amendment was applied in combination, the maximum value of the decrease of Cd content in edible parts of crops was between the maximum value of single application of two kinds of amendments, indicating that the organic-inorganic compound amendments could effectively inhibit the absorption of Cd by crops. In addition, organic-inorganic compound amendments have increased crop yield (Fig.3(c)).

The types of farmland soil in China are diverse, and the climatic conditions and tillage management levels are different, which leads to great differences in physical and chemical properties of different types of soil[29, 30]. As a result, the remediation effect of the same type of soil usually showed great uncertainty. Some results from pot studies cannot be applied in field environment [31] or the effect is poor after application.

Therefore, relevant work should be carried out continuously from the following aspects in the future:

(1) As for the problems of excessive heavy metals in passivation materials [32] and non-point source pollution caused by excessive application, etc. [33], it is better to systematically analyze the environmental risks of various amendments from the perspective of safety promotion and utilization of amendments, to further standardize the screening method of passivating agent from scientific research to application, and to introduce the environmental risk assessment into the impact assessment after the application of amendments.

(2) Since pot experiments were mostly carried out in controlled and semi-controlled environments,
while the spatial and temporal variability of soil moisture and nutrient content in the field, as well as root density and root depth of different plants is relatively large [34,35], so results from pot experiments were difficult to be used to estimate Cd uptake by crops under field conditions [36]. Therefore, it is necessary to enlarge the research results of combination between low-accumulation crop and amendment to the field test, and the changes of soil properties, soil fertility, soil health quality, and Cd content in crops after the application of different types of amendments should be followed up for several consecutive seasons, so as to obtain the continuous remediation effect of the combination method, to provide theoretical and practical reference for the large-scale extension of restoration scheme.

(3) On the basis of the above studies, the effectiveness and adaptability of different types of low-accumulation crops to heavy metal passivation and the compliance of crops under long-term planting conditions should be systematically studied for soils with different types and different levels of Cd pollution. Besides, a guidance database for low accumulation crop should be established for soils with different properties and Cd risks to provide technical support for effective and standardized remediation of Cd contaminated soils.

(4) Although morphology of heavy metal can be changed by passivating remediation technology, the total amount of heavy metal does not change. Whether the application of amendment can guarantee the long-term safe production of low-accumulation crops remains to be verified. In order to achieve the synchronization of heavy metal reduction and safe production of agricultural products, low accumulation crops and super accumulative plants can also be interplanted or rotated.

5. Conclusions

The combination of low accumulation crop with chemical amendment can effectively reduce the available Cd in soil and the content of Cd in agricultural products, but the application effect is quite different in soil with different Cd risk. In general, planting low-accumulation crops alone in soils with low-Cd risk can bring agricultural products up to standard. For soils with midium-Cd risk, the available Cd in soil and the content of Cd in edible parts of crops significantly decreased, and the average compliance rate of agricultural products was more than 90%, after the combined application of low-accumulation crops and amendments, indicating that the combined treatment of low-accumulation crops and amendment is more suitable for the restoration of soils with medium-Cd risk. Only 17% of the agricultural products reached the standard after the combined remediation of soils with high-Cd risk, indicating that the combined remediation method is not applicable in this case. Therefore, in the process of remediation of farmland contaminated by Cd, appropriate amendment should be selected according to the risk classification of heavy metals, cultivated land conditions and local economic factors, to formulate a scientific and objective remediation scheme for heavy metal pollution in farmland.

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