Remediation Effect of Aluminum Containing Compound Soil Conditioners on Cadmium and Plumbum Contaminated Soil and Vegetables

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Keywords: Zero-valent aluminium powder, Soil conditioner, Vegetable, Soil, Heavy metal, Contamination of Cadmium (Cd) and Plumbum (Pb).

Abstract. Some kinds of compound soil conditioners were made from zero-valent aluminium, sulfur and calcium hydroxide according to a certain weight proportion of those materials. And a soil-pot trial was conducted to test effects of using them on vegetable yield, contents of cadmium and plumbum in vegetable, and the available contents in soils. Results indicate that, on the basis of “Pollution soil + chemical fertilizer” (i.e., treatment of “PS+CF”), it would increase vegetable’s yield, and decrease the contents of cadmium and plumbum of vegetable, enhance soil’s pH, but reduce the available contents of Cd and Pb in soils to a certain extent by using those soil conditioners (i.e., treatment of “PS+CF+Al₁”, “PS+CF+Al₂”, “PS+CF+Al₃” and “PS+CF+Al₄”). Among them, the effects of treatment “PS+CF+Al₄” were relatively the best, and has a good application prospect in the field of soils, which were suffered from the contaminated by heavy metals Cd or Pb.

Introduction

With the acceleration of Chinese industrialization process and the rapid advancement of urbanization, the problem of heavy metal pollution in farmland soil in China has become increasingly prominent. According to the “National survey of soil pollution status of China” (April 17, 2014), the over-standard rate of spot soil pollution in China was 19.4%, of which the light-to-moderate pollution sites account for 94.3% of the total excess points; The types of soil pollution were mainly 8 heavy metals such as cadmium, mercury, arsenic, copper, plumbum, chromium, zinc, and nickel, and their over-scale points account for 82.8% of all over-target points. Due to heavy metal contamination of farmland soil, the problem of heavy metal pollution in agricultural products produced on contaminated farmland has been becoming increasingly prominent. And it has been reported that the excessive levels of heavy metals in Chinese vegetable soils were heavier than those in grain field.

At present, the main passivation materials used for in-situ passivation of heavy metals in soils include: phosphorus-containing materials, organic materials, silicon-calcium materials, clay minerals, metal oxides, biochar, and new materials. The use of zero-valent aluminum powders to passivate the activity of heavy metals in soil is still rare. Aluminum is the most abundant metal element on the earth and its sources are extensive, and it has the advantages of abundant resources, wide sources, low cost, and fast reaction speed. At the same time, since reducing aluminum is a porous substance, it has a high surface activity and can adsorb a variety of pollutants; and it is an active metal. The hydroxide colloid produced during the corrosion oxidation process has a strong adsorption effect and can effectively adsorb pollutants in the environmental media. However, there are few reports on the use of reducing aluminum to treat environmental pollutants. 
This study attempted to prepare some compound soil conditioners with reduced aluminum powder, sulfur and calcium hydroxide at a certain mass ratio, and applied them to the in-situ passivation and repair of farmland soils that were contaminated by heavy metals. In order to test the effects of those soil conditioners on the yield of vegetables, the contents of Cd and Pb in vegetables, and the contents of heavy metal Cd and Pb in soil, an experiment was conducted with soil-pot trial and 2 stubbles of vegetable including Chinese cabbage and water spinach as test crops. The results would provide new heavy metal passivation materials for the treatment of heavy metal polluted farmland.

Materials and Methods

The experiment was set up in the simulation room of the Institute of Soil and Fertilizers, Fujian Academy of Agricultural Sciences. The test soil was collected from the Fujian Science and Observation Station for Arable Land Conservation of Xitou Village, Baisha Town, Minhou County, Fujian Province. The soil was unpolluted paddy soil, and the parent material was hilly red soil, and the soil type was Huangnitian. In order to study the decontamination effect of heavy metal detergency agents, artificial addition of exogenous heavy metals were used to prepare experimental soils contaminated by heavy metals. The level of heavy metal pollution was designed to the level of "Soil Environmental Quality Standard" (GB15618-1995) (ie, Cd ≥ 1 mg·kg⁻¹, Pb ≥ 500 mg·kg⁻¹), among which Cd uses analytical reagent Cd(NO₃)₂·4H₂O, Pb uses analytical reagent Pb(NO₃)₂.

Seven trials were designed for the treatment, namely: (1) Clean soil (without addition of exogenous heavy metals Cd and Pb); (2) Contaminated soil (addition of exogenous heavy metals Cd 1 mg·kg⁻¹ soil, Pb 500 mg·kg⁻¹ soil, the same below); (3) Contaminated soil + chemical fertilizer (N 180 kg·hm⁻², and P₂O₅ 72kg·hm⁻², K₂O 90 kg·hm⁻² for each stubble vegetable, the same hereinafter); (4) Contaminated soil + fertilizer + aluminum-containing soil conditioner 1 (225 kg·hm⁻² per vegetable); (5) Contaminated soil + fertilizer + aluminum-containing soil conditioner 2 (255 kg·hm⁻² per vegetable); (6) Contaminated soil + chemical fertilizer + aluminum-containing soil conditioner 3 (each vegetable 172 5 kg·hm⁻²); (7) Contaminated soil + chemical fertilizer + aluminum-containing soil conditioner 4 (each vegetable 175 5 kg·hm⁻²). They are represented by seven codes, i.e.: CS, PS, PS+CF, PS+CF+Al₁, PS+CF+Al₂, PS+CF+Al₃, and PS+CF+Al₄. Each treatment was repeated 3 times. The test nitrogen fertilizer was urea (N46%), the phosphate fertilizer was monoammonium phosphate (N10%, P₂O₅50%), the potash fertilizer was potassium chloride (K₂O60%), and the reduced aluminum powder (passed 200 mesh sieve) was provided by Gongyi Dafa Metallurgical charging Co., Ltd. Sulfur and calcium hydroxide are analytical reagents, provided by Xitou Chemical Co., Ltd. of Shantou City. The chemical fertilizers and iron-containing soil conditioners used for each treatment were mixed well with the soil samples before sowing. The first vegetable was Chinese cabbage (Brassica campestris L. ssp., Qingjiang cabbage), which was planted on November 2, 2015 and harvested on January 28, 2016. The second crop was a spinach (Ipomoea aquatica Forsk, Thailand double axe) sowed on April 24, 2016 and harvested on August 16, 2016. At the time of harvest, the yield of vegetables was measured, and the contents of Cd and Pb in vegetables were measured by sampling, and the contents of available Cd and available Pb in the soil after harvesting.

The basic physicochemical properties of soil were determined by routine analysis of soil agrochemical methods[11]. The available heavy metals Cd and Pb in soil were determined by DTPA extraction-atomic absorption spectrophotometry[12]. Test data processing using Microsoft Excel-2003 office software, statistical analysis using SPSS11.0 statistical software.

Results and Analysis

Effects of Aluminum-containing Compound Soil Conditioners on the Biological Productivity of Vegetables

Results (Table 1) showed that on the basis of conventional application of chemical fertilizer treatment (PS+CF) to contaminated soils, several aluminum-containing soil conditioners were added (ie,
PS+CF+Al1, PS+CF+Al2, PS+CF+Al3 and PS+CF+Al4) could increase the biomass production of vegetables to a certain extent. Compared with PS+CF treatment, the biomass yields of the edible parts of the 1st and 2nd stubble vegetables were respectively different. It increased by 6.84%~35.67% and 12.45%~207.18%, and the average increase rate of the total output of two crops of vegetables was 7.65%~60.20%. Among them, treatments of PS+CF+Al3 and PS+CF+Al4 had relatively higher biological output. The total yield of 2 vegetables reached at 653.63 g·pot⁻¹ and 540.53 g·pot⁻¹, respectively, while the biomass yield of non-fertilized soil (PS) in the contaminated soil was the lowest, only 323.88 g·pot⁻¹. The treatment of CS also had a relatively lower yield. The results of variance analysis showed that the effect of improving total biomass of 2 vegetables was relatively better with PS+CF+Al3 and PS+CF+Al4, and it was significantly (P<0.01) better than other treatments. However, the difference between those two treatments was not significant (P>0.05); the PS+CF+Al2 and PS+CF+Al1 treatments are also significantly better than the PS and CS treatments. But the difference among treatments of PS+CF+Al2, PS+CF+Al1 and PS+CF was not significant.

Table 1. Effect of applying compound soil conditioners containing zero-valent aluminium on vegetable yield.

| Treatment | Chinese cabbage Yield/ g·pot⁻¹ | % | Water spinach Yield/ g·pot⁻¹ | % | Total Yield/ g·pot⁻¹ | % |
|-----------|-------------------------------|---|-------------------------------|---|----------------------|---|
| CS        | 296.67Bde                    | 8.74 | 81.41Bbcd                    | 39.53 | 378.08Dcd           | 16.74 |
| PS        | 272.82Be                     | -21.98 | 51.06Be                      | -12.49 | 323.88Dd            | -20.62 |
| PS+CF     | 349.66ABcde                  | 28.17 | 58.35Bde                     | 14.28 | 408.01Cde           | 25.98 |
| PS+CF+Al1 | 373.59ABbcd                  | 36.94 | 65.61Bbcde                   | 28.50 | 439.21Cbc           | 35.61 |
| PS+CF+Al2 | 382.04ABabcde                | 40.84 | 87.60Bbd                     | 71.56 | 471.84BDbc          | 45.68 |
| PS+CF+Al3 | 474.39Aa                     | 73.89 | 179.24Aa                     | 50.12 | 653.63Aa            | 60.20 |
| PS+CF+Al4 | 474.39Aa                     | 73.89 | 179.24Aa                     | 50.12 | 653.63Aa            | 60.20 |

Note: Different capital or small letters after data stand for very significant difference (P<0.01) or significant difference (P<0.05) in above table, the same below.

Effect of Several Aluminum-containing Soil Conditioners on the Contents of Cd and Pb in Vegetables

The test results (Table 2) showed that on the basis of conventional application of chemical fertilizer treatment (PS+CF) to contaminated soil, several aluminum-containing soil conditioners were added (ie, PS+CF+Al1, PS+CF+Al2, PS+CF+Al3 and PS+CF+Al4) could reduce the content of Cd and Pb in vegetables to a certain extent. Compared with PS+CF treatment, the content of Cd in both stubble vegetables was decreased by 12.46%~71.95% and 15.46%~74.69%, respectively. The decrease of Pb content in vegetables was 1.55%~18.77% and 6.98%~63.36%, respectively. Among them, PS+CF+Al4 and PS+CF+Al3 had a better effects on the decreasing of Cd and Pb in vegetables. The decreasing rates of Cd content in Chinese cabbage and water spinach of both treatments were 71.95% and 15.46%, respectively. And the decreasing rates of Pb were 18.77% and 4.36%, respectively. While the contents of Cd and Pb in both stubble of vegetables were highest in non-fertilized and contaminated soils treatment (PS). On the contrary, the contents of Cd and Pb in both stubble of vegetables were the lowest in clean and not fertilized soil treatment (CS). The results of variance analysis showed that: (1) The effect of decreasing the Cd content of Pakchoi cabbage was best with PS+CF+Al4, and it was significantly (P<0.01) better than that of PS, PS+CF, PS+CF+Al1 and PS+CF+Al2. And it was also significantly (P<0.05) better than that of PS+CF+Al3, but no significant difference with CS treatment. PS+CF+Al3 treatment was also extremely significant (P<0.01) superior to PS, PS+CF, and PS+CF+Al2 treatments, but the difference from PS+CF+Al1 treatments was not significant. The effect of reducing the content of Pb in the Chinese cabbage was not significant except for the clean soil treatment (CS). (2) Treatment of PS+CF+Al4 had a significantly (P<0.01) better effect on decreasing Cd content in water spinach than other treatments(except CS treatment). And PS+CF+Al3 treatment had a significantly (P<0.01) better
effect than PS, PS+CF, and PS+CF+Al\textsubscript{1} treatments, but it was not significantly different from PS+CF+Al\textsubscript{2} treatment. The reduction effect of Pb content in water spinach was better with PS+CF+Al\textsubscript{1} and PS+CF+Al\textsubscript{3}, and it was significantly ($P<0.01$) better than other treatments (except for CS treatment). However, the difference between the two treatments was not significant. PS+CF+Al\textsubscript{2} treatment was also significantly ($P<0.01$) better than PS and PS+CF treatment, but it was no significant difference from PS+CF+Al\textsubscript{1} treatment.

Table 2. Effect of applying compound soil conditioners containing zero-valent aluminium on contents of cadmium and plumbum in vegetable.

| Treatment       | Chinese cabbage | Water spinach |
|-----------------|-----------------|---------------|
|                 | Cd              | Pb            | Cd              | Pb              |
|                 | Content/ mg kg\textsuperscript{-1} | %  | Content/ mg kg\textsuperscript{-1} | %  | Content/ mg kg\textsuperscript{-1} | %  |
| CS              | 0.17Ee          | -92.11        | 0.08Bb          | -95.65          | 0.07Gf          | -93.5 |
| PS              | 2.13Aa          | 0.2           | 2.12Aa          | 13.94           | 1.09Aa          | 8.14 |
| PS+CF           | 2.13Aa          | \             | 1.86Aa          | \               | 1.01Aa          | \    |
| PS+CF+Al\textsubscript{1} | 1.83ABCabc | -13.8         | 1.77Aa          | -4.62           | 0.85Bb          | -15.46 |
| PS+CF+Al\textsubscript{2} | 1.86ABCa | -12.46        | 1.83Aa          | -1.55           | 0.65CDc          | -34.99 |
| PS+CF+Al\textsubscript{3} | 1.23BCDabc | -42.28        | 1.78Aa          | -3.46           | 0.60DEc          | -40.25 |
| PS+CF+Al\textsubscript{4} | 0.60DEde | -71.95        | 1.51Aa          | -18.77          | 0.25FGe          | -74.69 |

The Effect of Aluminum-containing Compound Soil Conditioners on Soil pH and Available Cd, Pb after Harvest

The results of the analysis of potted soil samples after planting 2 vegetables (Table 3) showed that: based on the conventional application of chemical fertilizer treatment to the contaminated soil(PS+CF), the addition of different soil conditioners could improve pH and reduce the content of available Cd and Pb in soil to a certain extent. The soil pH was increased by 4.01%~31.44% and the content of available Cd and Pb in soil was decreased by 7.07%~13.79% and 15.09%~32.42%, respectively. Among them, two treatments such as PS+CF+Al\textsubscript{4} and PS+CF+Al\textsubscript{3} had relatively best effects on improving soil pH and reducing soil available Cd and Pb content. The results of variance analysis showed that: (1) The treatments of PS+CF+Al\textsubscript{3} and PS+CF+Al\textsubscript{4} has very significant ($P<0.01$) effect on improving soil pH than others, but the difference between them was not significant. And the difference between other treatments was not significant. (2) The effect of clean soil (CS) treatment on the decreasing available Cd content in the soil was very significantly ($P<0.01$) better than that of other treatments; and the effective Cd content of the contaminated soil treatment (PS) was significantly higher than that of other treatments. Other treatments, such as PS+CF+Al\textsubscript{4}, PS+CF+Al\textsubscript{3}, and PS+CF+Al\textsubscript{2}, could significantly ($P<0.05$) decrease soil available Cd content compared to PS+CF treatment. (3) PS+CF+Al\textsubscript{4} treatment was relatively better for reducing the available Pb content in soil, and could be significantly ($P<0.01$) better than PS+CF and PS treatments, but the difference from PS+CF+Al\textsubscript{3}, PS+CF+Al\textsubscript{2} and PS+CF+Al\textsubscript{1} treatments was not significant; PS+CF+Al\textsubscript{1} and PS+CF+Al\textsubscript{2} treatments were also significantly ($P<0.05$) better than PS+CF and PS treatments, but The difference from PS+CF+Al\textsubscript{3} treatments was not significant. However, soil available Pb content in clean soil (CS) was significantly lower ($P<0.01$) than other treatments.

Table 3. Effect of applying compound soil conditioners containing zero-valent aluminium on pH, contents of available cadmium and plumbum in soil after harvest of vegetables.

| Treatment | pH | Available Cd | Available Pb |
|-----------|----|--------------|--------------|
|           | Value | % | % | Content/ mg kg\textsuperscript{-1} | %  | % | Content/ mg kg\textsuperscript{-1} | %  | % | |
| CS        | 4.14Cc | -6.54 | -6.05 | 0.01De | -99.42 | -99.29 | 1.63Ee | -99.47 | -99.45 | |
| PS        | 4.43BCc | \ | 0.53 | 0.95Aa | \ | 22.59 | 307.55Aa | \ | 3.65 | |
| PS+CF    | 4.41Cc | -0.53 | \ | 0.77Bb | -18.43 | \ | 296.71ABa | -3.52 | \ | |
Conclusions and Discussions

**Effect of Using Aluminum-containing Soil Conditioners on Vegetable Yield**

At present, there have not reports on the effect of applying aluminum-containing soil conditioners on the yield of vegetables grown in soils contaminated by heavy metals. The results of our experiment showed that: based on the conventional application of chemical fertilizer treatment to contaminated soil (PS+CF), several aluminum-containing soil conditioners were added could increase the biomass production of vegetables to a certain extent. Compared with PS+CF treatment, the yields of Chinese cabbage and Water spinach were increased by 6.84%~35.67% and 12.45%~207.18%, respectively. The average increase rate of the total output of two crops of vegetables was 7.65%~60.20%. Among them, PS+CF+Al$_3$ and PS+CF+Al$_4$ treatments had relatively higher biological outputs of vegetables, 2 stubble of vegetables reached at 653.63 g·pot$^{-1}$ and 540.53 g·pot$^{-1}$, respectively. The addition of several aluminum-containing soil conditioners could increase the biological productivity of vegetables to some extent. The reason is that using those soil conditioners could form a variety of insoluble compounds with active cadmium and lead ions in the soils, and reduce the contents of Cd and Pb in the soils to a certain extent. Thus it alleviates the excess of heavy metal elements in the soil which inhibit the growth of vegetables and improves the yields of vegetables.

**Effect of Using Aluminum-containing Soil Conditioners on Heavy Metal Content in Vegetable-soil System**

At present, there is no report on the treatment of heavy metal pollution in agricultural soils with aluminum-containing soil conditioners. However, studies on the repair of heavy metal-contaminated soil with zero-valent iron have been reported$^{[13,14]}$. The physical and chemical properties of zero-valent aluminum and zero-valent iron are similar, and the passivation mechanisms for the reduction and adsorption of heavy metal pollutants in environmental media are similar. What is even more remarkable is that zero-valent aluminum has unique advantages over zero-valent iron: First, aluminum is more abundant in nature than iron, so its sources are more extensive$^{[8]}$. Second, the reduction potential of zero-valence aluminum is $E_0$=-1.662 V, and the reduction potential of zero-valent iron is $E_0$=-0.44 V, so zero-valent aluminum has a stronger electron transfer ability, that is, its metal reducibility is stronger$^{[10]}$. At present, there is less research on zero-valent aluminum, but because of its wide range of sources, it has a stronger metal activity, can be used as a stronger reducing agent, and has great application prospects in the treatment of waste water pollution, even at home and abroad. One of the most promising material governance technologies is considered$^{[8]}$. However, there is no report on the application of aluminum-containing soil conditioners on vegetable fields. The results of our experiment showed that: (1) Based on the conventional application of chemical fertilizer treatment to contaminated soil (PS+CF), several aluminum-containing soil conditioners were added could reduce the content of Cd and Pb in vegetables to a certain extent. Compared with the PS+CF treatment, the decreasing amplitude of the content of Cd in the first stubble and second stubble vegetables was 12.46%~71.95% and 15.46%~74.69% respectively, and the decrease of Pb content was 1.55%~18.77% and 6.98%~63.36%, respectively. Among them, the reducing effect of PS+CF+Al$_4$ and PS+CF+Al$_3$ on the content of Cd and Pb of vegetable weight was relatively better. The decrease of Cd content in the first stubble and second stubble vegetables were 71.95% and 42.28%, 74.69% and 40.25%, respectively; and that of Pb were 18.77% and 4.36%, 63.36% and 61.92%, respectively.(2) On the basis of conventional fertilizer treatment (PS+CF), using those soil conditioners could improve the soil pH after the harvest of vegetables to a certain extent, and the increase rate was 4.01%~31.44%, and the content of available Cd and Pb in soil was reduced, and the
decrease rates were 7.07%~13.79% and 15.09%~32.42% respectively. Among them, treatments of PS+CF+Al4 and PS+CF+Al3 had the best effect on improving soil pH and decreasing soil available Cd and Pb content.

To sum up, adding "aluminum containing heavy metal passivating agent 4" in farmland with heavy metal Cd and Pb content in soil can not only significantly increase the biological yield of vegetables, reduce the content of Cd and Pb in vegetable heavy metals, but also obviously reduce the content of available Cd and Pb in the soil. Therefore, the aluminum containing soil conditioner has a certain promotion and application prospect in the farmland with high content of heavy metal Cd and Pb in soil, but the results of this experiment are only obtained through the greenhouse pot experiment, and the field test should be further verified.

Acknowledgement
This research was financially supported by National science and technology support program (2015BAD05B01-05), Fujian province belongs to the basic scientific research project of public welfare (2017R1022-1, 2018R1022-4); The scientific and technological innovation team of the Fujian Academy of Agricultural Sciences (STT2017-2-10); and The research center of Fujian provincial land cultivation engineering technology.

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