

## Article

# Enhanced Phytoremediation for Trace-Metal-Polluted Farmland with *Hibiscus cannabinus*–*Sedum plumbizincicola* Rotation: A Case Study in Hunan, China

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**Abstract:** Trace metal pollution in farmland threatens the health of both crops and humans. Restoring these polluted farmlands safely and utilizing them to elevate farmers' incomes are extremely needed. Phytoremediation is a promising method for metal extracting but its popularization is limited by both its low efficiency and the low economic value of the plants used. Herein, a field study was conducted to investigate the potential of using a rotation with the hyperaccumulator of *Sedum plumbizincicola* and kenaf (*Hibiscus cannabinus*) for combined heavy-metal-contaminated farmland remediation. Results showed that the kenaf obtained an aerial biomass of up to 21 Mg ha<sup>-1</sup> under combined heavy metal contaminations, which was significantly higher than that for *S. plumbizincicola* (<8 Mg ha<sup>-1</sup>). However, the concentrations of Cd, Cu, Pb, and Zn in *S. plumbizincicola* were at least 100, 2, 8, and 75 fold higher than that for kenaf, respectively. The removal of Cd, Pb, and Zn for *S. plumbizincicola* can be more than 3800, 720, and 104,347 g ha<sup>-1</sup>, which was at least 38, 3, and 27 times higher than that for kenaf, respectively. Finally, the removal of Cd, Cu, Pb, and Zn by rotation of the two crops was increased by 7.88%, 126%, 33.5%, and 4.39%, respectively, compared with the *S. plumbizincicola* monoculture. Hence, the rotation with kenaf and *S. plumbizincicola* can not only remove more heavy metals from the contaminated soil and accelerate the phytoremediation pace, but also can supply a large number of raw materials for industrial applications.

**Keywords:** cadmium; copper; *Hibiscus cannabinus*; lead; soil pollution; zinc

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## 1. Introduction

Heavy metal (HM) pollution has long been a focus problem in the world owing to its serious toxicity to both environment and organisms [1–3]. Under the dual action of human activity factors and natural factors, HM pollution in soil and water is increasingly serious [4,5] and severely restricts the sustainable development of agriculture in China. Considering the increasing population but decreasing farmland area, remediating and reusing HM-polluted lands is becoming more and more necessary and urgent [6]. Among the remediation methods, phytoremediation is recognized as the most promising one for its unique advantages: in situ applicability, easy implementation, non-invasive, low-cost, and solar-driven [7–9]. However, every coin has two sides. Phytoremediation also has its shortcomings. On the one hand, phytoremediation always presents a discouragingly low remediation efficiency [10]. On the other hand, most of the plants used for phytoremediation are of little economic value, which cannot elevate the passion of farmers for cultivating such plants. All the above have limited the popularity of phytoremediation. Hence, the selection of a plant for remediation is often crucial and open to question.

The genus *Sedum* L. possesses the most species (about 420 species) members among the family of Crassulaceae, which can endure harsh environments, such as cold and hot

temperatures and dry conditions [11,12]. Among the species, the *S. plumbizincicola* found in Cd/Zn mining areas in Southeast China, is well known for its excellent performance in accumulating, translocating, and tolerating high concentrations of HMs, particularly for Cd and Zn, and consequently has become one of the most popular hyperaccumulators in trace metal phytoremediation [13–15]. It is estimated that the Cd and Zn concentrations can be up to 400 and 10,000 mg kg<sup>-1</sup>, respectively, in *S. plumbizincicola* [11]. However, the biomass of *S. plumbizincicola* is limited, which greatly lowers the remediation efficiency for HM pollution. On the other hand, the economic value of *S. plumbizincicola* is also much lower. Hence, seeking HM-tolerant and high-economic-value crops for phytoremediation is a better way to restore polluted farmlands.

Bast fiber crops, such as kenaf (*Hibiscus cannabinus* L.), flax (*Linum usitatissimum* L.), and hemp (*Cannabis sativa* L.), are traditional crops with a long cultivation history. However, they have been applied to food, medicine, biodegradable composites, and building industries, in addition to textiles, and show great economic value [16–18]. For example, Kong et al. found that the incorporation of kenaf straw core or kenaf fiber can increase the viscosity recovery and thickness shape retention of geopolymer and improve the satisfactory performance of printed specimens [19]. Apart from their multiple uses, these crops can grow fast with huge biomass and can be promising candidates for energy crops [20–22]. By theoretical calculation, it is thought that kenaf can produce 61.4 gals of biofuels per ton of biomass [23]. It has been reported that the dry stem material of kenaf can be up to 30 Mg ha<sup>-1</sup> year<sup>-1</sup> under suitable conditions [24]. Furthermore, bast crops have been proven to be tolerant to various HMs, such as Cd, Pb, Zn, Sb, Cr, and Ni [25–28]. A previous study found that the Cd and Pb concentrations in kenaf seedlings can reach 355 mg kg<sup>-1</sup> and 606 mg kg<sup>-1</sup>, respectively, under pot experiments with artificially contaminated soil [26]. Despite lower bioconcentration factors compared with hyperaccumulators, given their huge biomass, bast fiber crops are still suitable to be used for phytoremediation, especially in fields no longer suitable for cultivating food crops.

Since *S. plumbizincicola* can resist low temperatures, it is usually planted in November in Hunan Province, China. On the contrary, kenaf is tolerant of high temperatures and is usually cultivated in May. Hence, a rotation with these two crops can make full use of farmland during the year and may improve remediation efficiency. Considering the characteristics of *S. plumbizincicola* (the excellent ability to extract heavy metals) and kenaf (huge biomass and multiple uses), the combined use of two crops for remediating HM-contaminated soil seems a good choice. Therefore, a field rotation trial was conducted in a typical multi-HM-contaminated field with kenaf and *S. plumbizincicola* for phytoremediation. This study aims to: (i) discuss the response of both crops to the combined HM pollution; and (ii) clarify the potential of a kenaf–*S. plumbizincicola* rotation for phytoremediation. We hypothesized that the rotation of two crops can enhance the remediation efficiency of HM-polluted soil and the cultivation of *S. plumbizincicola* in rotation can increase the uptake of HM when followed by kenaf.

## 2. Materials and Methods

### 2.1. Experimental Site and Design

The experimental site (28°16'42" N, 113°55'21" E) was located at Yonghe Town, Liuyang City, Changsha City, Hunan, China, and has a typical subtropical monsoon humid climate with an annual average temperature of 16.7 °C and an annual average rainfall of 1500 mm. The site previously belonged to Qibaoshan Town, which is famous for its rich nonferrous metal resources, such as Zn, Cu, and Pb. However, the historic development of such resources without protecting the environment has led to serious HM pollution in a wide area of farmland. The soil from the experiment field featured combined HM pollution and was classified as having a loamy clay texture (clay 27.1%, silt 39.5%, and sand 33.4%). The physicochemical properties of the soil are given in Table 1. According to the standard *Risk control standard for soil contamination of agricultural land (GB 15618—2018)* in China, the soil was seriously contaminated with Cd, Pb, Cu, and Zn.

**Table 1.** The physicochemical properties of the soil in the experimental site.

Items	Soil Organic Matter	Total Nitrogen	Available P	Available K	Total Cd	Total Pb	Total Cu	Total Zn	pH
	g/kg				mg/kg				
Values	42.2	2000	27.6	208	5.10	1376	840	770	5.70
Critical Values <sup>1</sup>		-	-	-	0.30	90	150	200	5.5 < pH ≤ 6.5

<sup>1</sup> Soil pollution risk screening values of agricultural land according to the standard *Risk control standard for soil contamination of agricultural land (GB 15618—2018)* of China.

A randomized block plot experiment with three treatments was designed in this study: monoculture with *S. plumbizincicola* (abbreviation for treatment, Sedum); kenaf (Kenaf); and a rotation with *S. plumbizincicola* and kenaf (Sedum–Kenaf). Each treatment in this study was replicated four times. The plot was 4 m long and 1.5 m wide. There were ditches (width, 0.5 m) between plots for distinguishing different plots and irrigation and drainage. Before growing any plants, a base fertilizer (Hubei Great Harvest Fertilizer Co., Ltd., Wuhan City, China Compound Fertilizer, N-P<sub>2</sub>O<sub>5</sub>-K<sub>2</sub>O, 15-15-15, 500 kg/ha for each season) was broadcasted onto each plot and evenly incorporated into the surface soil. In addition, urea (Henan Jinkai Chemical Investment Holding Group Co., Ltd., Urea, N, 46%, 160 kg/ha for each season) was applied by broadcasting at the vigorous growing stage for both crops. The seedlings of *S. plumbizincicola* were purchased from the market and planted in November 2020 with a row space of 20 cm and a line space of 20 cm. Kenaf was sown after the harvest of *S. plumbizincicola* in May 2021. Particularly, the kenaf seeds were sown in the same positions as the roots of the *S. plumbizincicola* to investigate the effects of *S. plumbizincicola* on the metal accumulation in kenaf. During the experimental period, weeding and irrigation were applied when it was needed.

## 2.2. Sampling and Measurement

Soil samples were collected two times: before the experiment began, soil samples were collected for the analysis of basic physicochemical properties; and when the experiment was finished, soil samples were collected to determine the metal concentrations in different fractions. When the soil samples were brought to the lab, the soil was air-dried and ground for further analysis. Plant samples were collected at the anthesis stage (at this stage the biomass of both plants reaches the maximum) for both kenaf and *S. plumbizincicola*: firstly, the total fresh biomass of the plant (roots and the aboveground part) for each trial plot was recorded, then 2 kg of the biomass per plot was brought to the lab as one sample. After that, the plant samples were divided into roots and the aboveground part and dried in an oven. The dried plant samples were then ground into fine powder for HM determination.

Soil pH was measured in a 1:2.5 soil/water suspension with a lab pH meter [29]. The total N concentration of the soil was determined with the Kjeldhal method [30]. Soil available P was extracted with 0.5 M NaHCO<sub>3</sub> (pH 8.5) with a liquid-to-solid ratio of 20:1 and determined through a colorimetric method [26]. Soil available K was extracted with ammonium acetate (pH = 7.0, w:v, 1: 10) and determined with a flame photometer (Sherwood, M410) [29]. The soil organic matter was determined with the potassium dichromate combustion method [31]. The particle size distribution of the soil was measured by the pipette method following the procedure described by Lu [29] and the texture was classified based on the triangular chart of international soil texture. Total HM concentrations in the soil samples were determined with an inductively coupled plasma optical emission spectrometer (ICPE-9820, Shimadzu Corporation, Kyoto, Japan) after being digested by HF-HClO<sub>4</sub>-HNO<sub>3</sub> in a microwave reaction system (CEM MARS6 CLASSIC, CEM Corporation, Charlotte City, NC, USA) [6]. The modified European Community Bureau of Reference (BCR) method was used to sequentially extract HM fractions in the soil samples [32]. In brief: (1) 1 g of the soil sample and 40 mL acetic acid (0.11 M) were added into the centrifuge tube and shaken for 16 h at room temperature, then the extract was separated by

centrifugation and put through a membranes-aquo system (0.45  $\mu\text{m}$ ), and the final filtrate was used to determine the mild-acid-soluble fraction of metals. (2) An amount of 40 mL of hydroxylammonium chloride (0.1 M) was added to the residue from step 1 and shaken for 16 h at room temperature again, and the extract and the final filtrate were treated with the same procedure as described in Step 1. (3) Firstly, 10 mL of hydrogen peroxide (8.8 M) was added to the residue from Step 2, then the mixture was allowed to rest for 1 h at room temperature with occasional manual shaking, then, after that, the mixture was processed with a water bath ( $85 \pm 2$  °C) for another 1 h. Then the centrifuge tube was uncovered and the digestion volume was reduced to about 2–3 mL by continual heating. The above procedures were then repeated once again. When the mixture had cooled down to room temperature, 50 mL of ammonium acetate (1.0 M) was added to the mixture and shaken for 16 h at room temperature. The extract and the final filtrate were treated with the same procedure as described in Step 1. (4) The residue collected from Step 3 was digested with a mixture of aqua regia and HF and the digestion solution was used to determine the residual fraction of metals.

The powdery plant samples were digested with analytically pure nitric acid (*w:v* 1: 50) via a microwave reaction system (CEM MARS6 CLASSIC, USA), then the digested solutions were also used for HM concentration analysis with an inductively coupled plasma optical emission spectrometer (ICPE-9820, Japan).

### 2.3. Data Analysis

The translocation factor (TF) and bioconcentration factor (BCF) for each HM were calculated with the following equations [33]:

$$\text{TF} = \text{HM concentration in the aerial part} / \text{HM concentration in the root}$$

$$\text{BCF} = \text{HM concentration in plant tissue} / \text{HM concentration in soil}$$

All the data presented in tables or figures were the mean values of the four replications. The data obtained from the experiment were analyzed with Microsoft Excel 2016 (Microsoft, Redmond, DC, USA) and SPSS 22.0 (IBM, Chicago, IL, USA). The graphs were plotted with Origin 2021 (Origin Lab, Northampton, MA, USA). The mean values of the variables for each treatment were compared using Duncan's test at  $p \leq 5\%$ .

## 3. Results

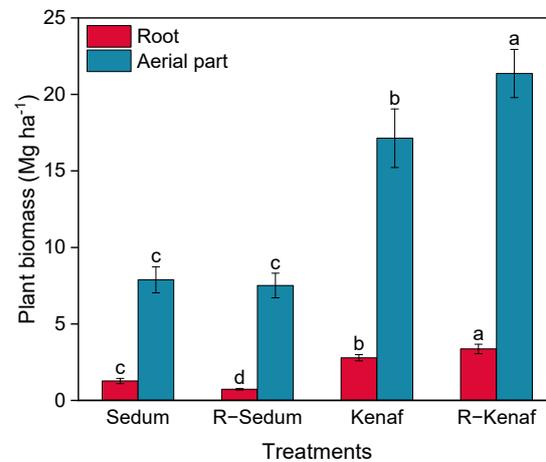
### 3.1. Plant Biomass under Combined HM Pollution

The results showed that the biomass of the two plants differed significantly for both the aerial part and the root (Figure 1). The average biomass of *S. plumbizincicola* (average value of Sedum and R-Sedum) under the combined HM contamination were 7700 and 1003 kg/ha for the aerial part and root, respectively. The average biomass of kenaf (average value of Kenaf and R-Kenaf) under the combined HM contamination was 19,256 and 3084 kg/ha for aerial part and root, respectively. Hence, growing kenaf can obtain at least twice the biomass of *S. plumbizincicola* on farmland contaminated with Cd, Pb, Cu, and Zn. No significant difference for the aerial part of *S. plumbizincicola* was found between Sedum and R-Sedum, however, the biomass of R-Kenaf was significantly higher than that of Kenaf. For the roots, the biomass of Sedum was notably higher than that of R-Sedum. However, the roots of R-Kenaf showed significantly higher biomass than that of Kenaf.

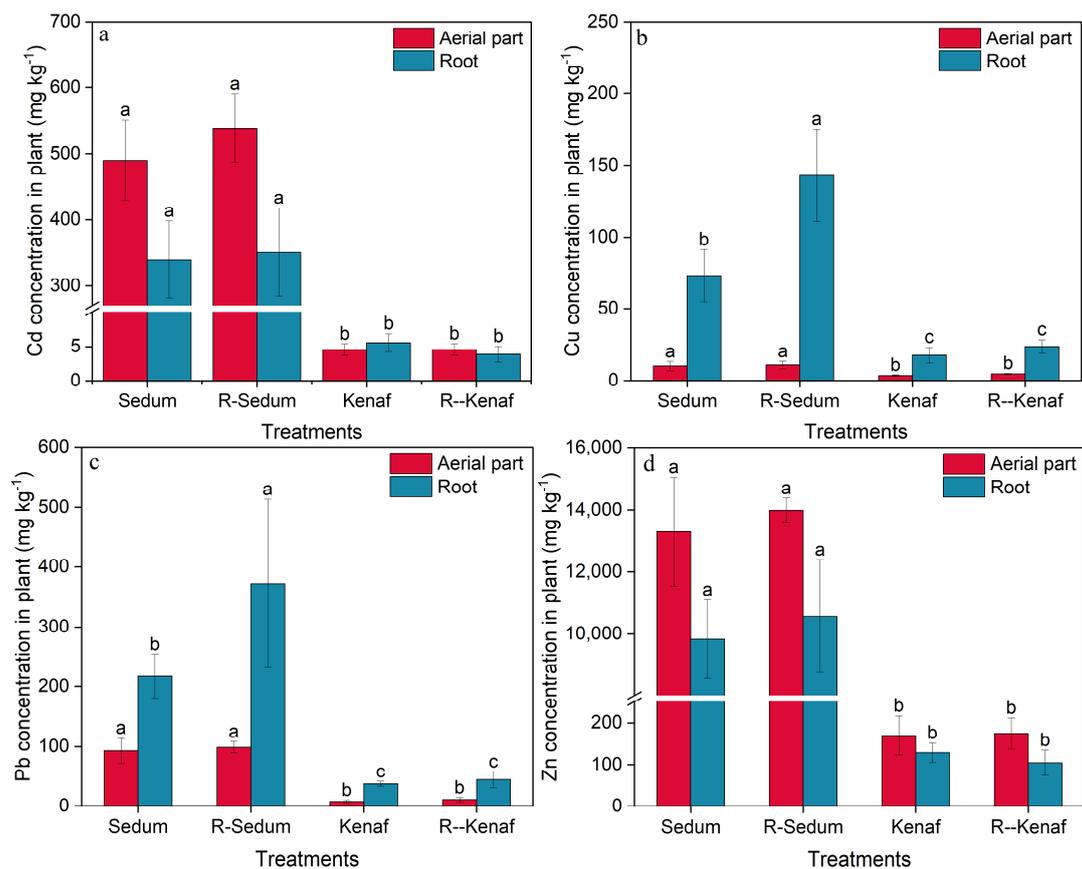
### 3.2. Heavy Metals in Plants

The HM concentrations in the aerial part and roots for both crops showed significant differences and the concentrations of the four heavy metals also differed greatly between treatments and crops (Figure 2). The concentrations of Cd in the aerial part and root of *S. plumbizincicola* were higher than 480 and 330 mg/kg, respectively, but were even lower than 6 mg/kg in the two parts of kenaf. Hence, the Cd concentration in *S. plumbizincicola* can

be as high as 80 times that in kenaf. However, no significant difference in Cd concentrations was shown between Sedum and R-Sedum or Kenaf and R-Kenaf.



**Figure 1.** The biomasses of kenaf and *S. plumbizincicola*. Letters above the columns for each variable indicate significant differences between treatments or crops. R–Sedum means the *S. plumbizincicola* in rotation with kenaf; R–Kenaf means the kenaf in rotation with *S. plumbizincicola*.



**Figure 2.** Metal concentrations of Cd (a), Cu (b), Pb (c), and Zn (d) in the aerial part and root for both kenaf and *S. plumbizincicola*. Letters above the columns for each variable indicate significant differences between treatments or crops. R–Sedum means the *S. plumbizincicola* in rotation with kenaf; R–Kenaf means the kenaf in rotation with *S. plumbizincicola*.

Unlike Cd, the concentrations of Cu were higher in the root for both crops compared with that in the aerial part. The Cu concentrations in *S. plumbizincicola* were far lower than

the Cd concentrations, but the Cu concentrations were much higher in kenaf than the Cd concentrations. However, *S. plumbizincicola* still took up more Cu than kenaf in both the aerial part and roots. There were no significant differences in Cu concentrations between Kenaf and R-Kenaf in both the aerial part and roots, but the Cu concentration in R-Sedum was significantly higher than that in Sedum in the root part.

Similar to Cu, the concentrations of Pb were also higher in the root part than that in the aerial part for both crops, and they were significantly higher in *S. plumbizincicola* than those in kenaf. No notable difference in Pb concentrations in the aerial part was shown between Sedum and R-Sedum, but the Pb concentration in the root part was notably higher for R-Sedum than that for Sedum. There were no marked differences in Pb concentrations between Kenaf and R-Kenaf.

The Zn concentrations showed a similar variation with Cd in the two crops and three treatments. However, the concentrations of Zn in both crops were far higher compared with those of Cd in the two crops. The Zn concentrations in *S. plumbizincicola* ranged from 13,296 mg kg<sup>-1</sup> to 13,993 mg kg<sup>-1</sup> and 9830 mg kg<sup>-1</sup> to 10,573 mg kg<sup>-1</sup> for the aerial part and roots, respectively. The Zn concentrations in kenaf were no more than 180 mg kg<sup>-1</sup>, which was also much higher than the Cd concentrations in kenaf. The Zn concentrations for the aerial part in *S. plumbizincicola* were more than 70 times higher than those in kenaf. There were no marked differences in Zn concentrations between Sedum and R-Sedum or Kenaf and R-Kenaf.

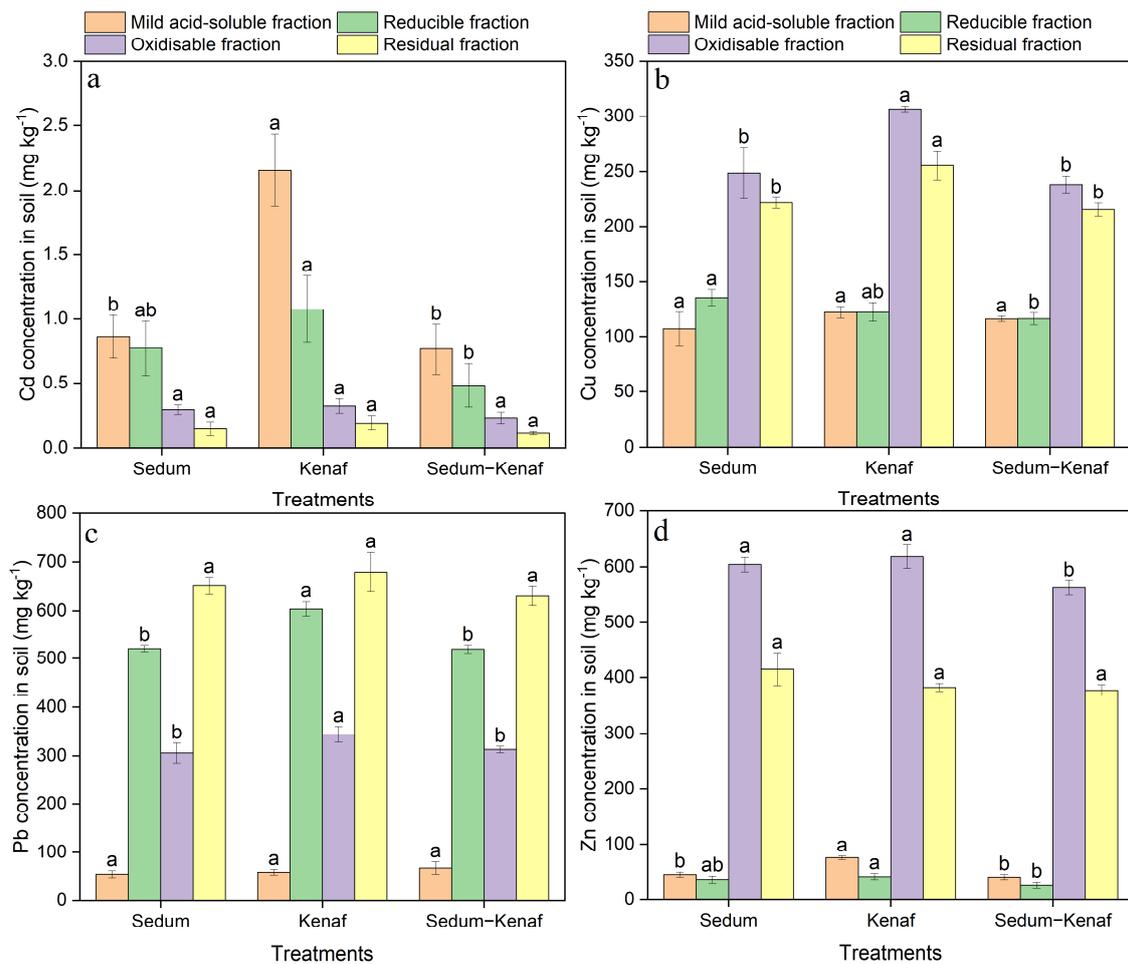
### 3.3. Heavy Metal Status in Soil

Soil metal concentrations of different fractions can be significantly affected by the crops and treatments (Figure 3). The Cd concentration of the mild-acid-soluble fraction with the Kenaf treatment was significantly higher than that for the Sedum and Sedum–Kenaf treatments. The Cd concentration of the reducible fraction for the Kenaf treatment was notably higher than that for the Sedum–Kenaf treatment but was only insignificantly higher than that for Sedum. For the Cd concentration of the oxidizable and residual fractions, no marked differences were found between the treatments. Furthermore, the Cd concentrations of the four fractions for Sedum were all higher than those for the Sedum–Kenaf treatment despite no notable difference between the two treatments.

Unlike Cd, the Cu concentrations of the mild-acid-soluble fraction differed insignificantly between treatments. The Cu concentration of the reducible fraction for Sedum showed no notable difference compared with that for the Kenaf treatment, but was significantly higher than that for Sedum–Kenaf. For the oxidizable and residual fractions, the Cu concentrations for the Kenaf treatment were markedly higher than those for the other two treatments, but no significant differences were shown between the Sedum and Sedum–Kenaf treatments.

The mild-acid-soluble fraction of Pb showed no significant difference between three the treatments. However, the reducible and oxidizable fractions of Pb for the Kenaf treatment were notably higher than those for the Sedum and Sedum–Kenaf treatments. No notable difference in the reducible and oxidizable fractions of Pb was found between the Sedum and Sedum–Kenaf treatments. The residual fraction of Pb also differed insignificantly between treatments but this fraction of Pb for the Kenaf treatment was slightly higher than those for the other treatments.

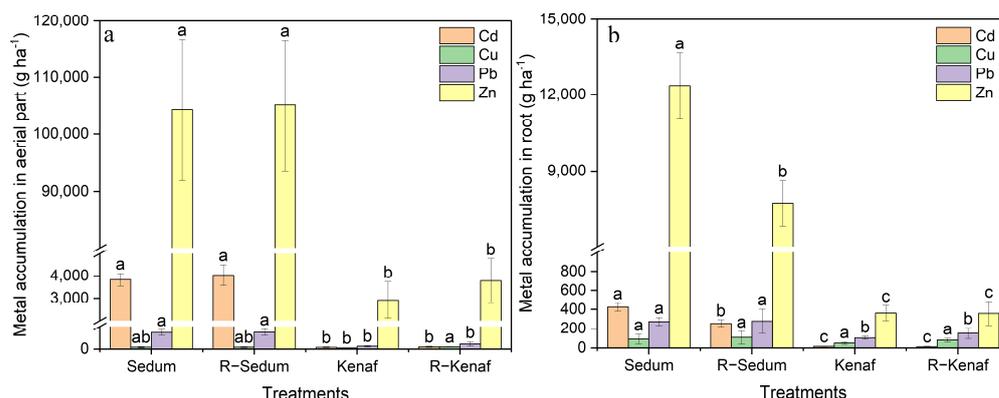
The mild-acid-soluble fraction of Zn for the Kenaf treatment was significantly higher than those for the Sedum and Sedum–Kenaf treatments. For the residual and oxidizable fractions of Zn, they were notably higher for the Kenaf treatment than those for the Sedum–Kenaf treatment but only slightly higher than that for the Sedum treatment. No significant difference was found for the residual fraction of Zn between treatments.



**Figure 3.** The soil metal concentrations of Cd (a), Cu (b), Pb (c), and Zn (d) in different fractions extracted with the BCR method at the end of the field experiment. Letters above the columns for each variable indicate significant differences between treatments or crops.

### 3.4. Heavy Metal Removal by Plants

The removal of HMs by *S. plumbizincicola* and kenaf was calculated as the outcome of the dry biomass of each plant multiplied by the according metal concentrations and is shown in Figure 4. The accumulation of Cd in the aerial parts showed no significant difference between treatments for the same crop but they were significantly higher for *S. plumbizincicola* (3832 g ha<sup>-1</sup> for Sedum and 4035 g ha<sup>-1</sup> for R-Sedum) than that for kenaf (78 g ha<sup>-1</sup> for Kenaf and 99 g ha<sup>-1</sup> for R-Kenaf). Similar to Cd in the aerial parts, the accumulation of Cu also differed indistinctively between treatments for *S. plumbizincicola*, however, it was notably higher for R-Kenaf than that for Kenaf. No significant differences were shown between the two crops for Cu accumulation in the aerial parts. For Pb in the aerial parts, the accumulation did not differ significantly between treatments for each crop but was notably higher for *S. plumbizincicola* than that for kenaf. The accumulation of Zn for both crops (with an average value of 104,746 g ha<sup>-1</sup> and 3354 g ha<sup>-1</sup> for *S. plumbizincicola* and kenaf, respectively) was far more than those of the other three metals. The accumulation of all the metal sorts in the aerial part for R-Sedum was higher than that for the Sedum treatment even though no significant differences were shown between treatments, and this phenomenon was also reflected between the kenaf treatments.



**Figure 4.** Metal accumulations in aerial part (a) and root (b) for kenaf and *S. plumbizincicola* under different treatments. Letters above the columns for each variable indicate significant differences between treatments or crops. R–Sedum means the *S. plumbizincicola* in rotation with kenaf; R–Kenaf means the kenaf in rotation with *S. plumbizincicola*.

The accumulation of four metals in the roots showed similar variations to those in the aerial parts. The accumulation of Cd in the roots of *S. plumbizincicola* was significantly higher than that of kenaf. Sedum accumulated a notably higher Cd than R-Sedum, but the Cd accumulation differed indistinctively between Kenaf and R-Kenaf. The accumulation of Cu in roots showed no notable difference between crops or treatments. The Pb accumulation differed indistinctively between treatments for the same crop but was significantly higher in *S. plumbizincicola* than in kenaf. *S. plumbizincicola* can accumulate a significantly higher amount of Zn than kenaf. The Cd accumulation in roots for Sedum were notably higher than that for R-Sedum but they differed indistinctively between Kenaf and R-Kenaf. The removal of all the metal sorts for R-Kenaf were higher than those for the Kenaf treatment even though no significant differences were shown between treatments. The removal of Pb and Cu by roots for R-Sedum was higher than that for the Sedum treatment even though no significant differences were shown between treatments. However, the removal of Cd and Zn for R-Sedum was notably lower than that for the Sedum treatment.

### 3.5. Translocation Factors (TF) and Bioconcentration Factors (BCF) for Each HM

The TF and BCF varied between treatments and crops (Table 2). The TFs for Sedum and R-Sedum were significantly higher than that for Kenaf and slightly higher than that for R-Kenaf. The TFs of Cu for kenaf from both the Kenaf and R-Kenaf treatments were notably higher than that for R-Sedum but showed no significant difference compared with that for the Sedum treatment. For Pb, the TF for Sedum was significantly higher than that for the other three treatments. However, no marked difference was found in the TFs of Zn between treatments. The BCFs of the four HMs in the aerial parts for *S. plumbizincicola* were significantly higher than those for kenaf. Furthermore, in the aerial part, the BCFs of the R-Kenaf treatment were slightly higher than those of the Kenaf treatment, regardless of the HM sort. The BCFs in the root varied similarly to the aerial part.

**Table 2.** Translocation factors and bioconcentration factors for HMs of two plants.

Items	Element	Sedum	R-Sedum	Kenaf	R-Kenaf
TF <sup>1</sup>	Cd	1.458 a <sup>3</sup>	1.568 a	0.875 b	1.218 ab
	Cu	0.164 a	0.089 b	0.214 a	0.197 a
	Pb	0.437 a	0.292 b	0.208 b	0.233 b
	Zn	1.362 a	1.352 a	1.368 a	1.732 a
BCF <sup>2</sup> -aerial part	Cd	111.29 b	254.02 a	1.207 c	3.280 c
	Cu	0.0136 a	0.0138 a	0.00524 b	0.00715 b
	Pb	0.0585 a	0.0601 a	0.00558 b	0.00759 b
	Zn	11.87 a	13.34 a	0.153 b	0.183 b

Table 2. Cont.

Items	Element	Sedum	R-Sedum	Kenaf	R-Kenaf
BCF-root	Cd	75.14 b	164.30 a	1.547 c	2.662 c
	Cu	0.0845 b	0.204 a	0.0258 b	0.0380 b
	Pb	0.129 b	0.250 a	0.0235 c	0.0322 c
	Zn	8.62 a	9.86 a	0.120 b	0.114 b

<sup>1</sup> Translocation factor; <sup>2</sup> Bioconcentration factor; <sup>3</sup> Different letters in each line indicate significant differences between treatments.

#### 4. Discussion

HM contaminations have seriously affected the normal use of farmlands, which not only reduces the utilization efficiency of limited land resources but also lowers farmers' incomes [34]. Hence, restoring contaminated farmlands with green techniques is urgent. Although phytoremediation has many merits, the low remediation efficiency and low economic value of most of hyperaccumulators hinder its application [10]. Thus, in this study, a remediation strategy with the rotation of *S. plumbizincicola* and kenaf was put forward. On the one hand, *S. plumbizincicola* has a great ability to concentrate HMs [11] and can remove metals at a relatively fast pace. On the other hand, kenaf has good tolerance to several HMs [26] and can obtain a huge amount of biomass, which can remove a certain amount of metals and be used for industrial applications, such as concrete reinforcement, fiber-reinforced polymer, decorative plate manufacture, energy production, textiles, and so on [20,35]. Therefore, it was expected that the rotation with two crops could improve the remediation efficiency and produce economic profits at the same time.

The results in the present study showed that the kenaf can obtain a huge aerial biomass (more than 21 Mg ha<sup>-1</sup>) on farmland heavily contaminated with complex HMs. However, under good conditions, kenaf can produce up to 30 Mg ha<sup>-1</sup> year<sup>-1</sup> of dry stem material [24]. This indicated that the growth of kenaf was restrained by the metal contamination. Owing to the gradual exhaustion of fossil resources and global climate change, bio-energy has attracted more and more attention [21,22,36], and the biomass or straw resources of crops will play an important role in the future [37]. It has been reported that kenaf stems can produce a heating value of 19.23 MJ/kg and can yield 106.8 gallons of ethanol per ton of dry biomass [22]. Since the current price of ethanol is about 1000 USD/Mg, cultivating kenaf on contaminated farmland can achieve a production value of 7000 USD/ha. Hence, if the production cost of ethanol can be further reduced, soil decontamination and economic profits can be obtained together. To cope with global climate change, greenhouse gas (GHG) emissions must be controlled [38]. The Chinese government has set a goal of 'peaking carbon dioxide emissions by 2030 and striving to achieve carbon neutrality by 2060' [39]. Phyto-sequestration seems to be an efficient way to achieve this goal because plants consume lots of CO<sub>2</sub> to produce biomass [40,41]. For this aspect, a huge amount of kenaf biomass can serve as a temporary carbon pool. Furthermore, when kenaf biomass is applied to composite materials, reinforced concrete, and so on, this temporary carbon pool will turn into a long-term carbon pool. Hence, as a crop featuring a high biomass, fast growth rate, and strong tolerance to HMs, kenaf should be a good choice for farmland that is no longer suitable for food production. Compared with kenaf, the biomass of *S. plumbizincicola* (less than 8 Mg ha<sup>-1</sup>) was much lower. However, the metal removal abilities of *S. plumbizincicola* are somewhat higher than those of kenaf. Results in this study showed that the concentrations of Cd, Cu, Pb, and Zn in *S. plumbizincicola* was at least 100, 2, 8, and 75 times higher than that for kenaf, respectively. Hence, despite the relatively small biomass, the removal of Cd, Pb, and Zn can be more than 3800, 720, and 104,347 g ha<sup>-1</sup>, which is at least 38, 3, and 27 times higher than that for kenaf. Interestingly, the metal removals for kenaf in rotation were slightly higher than those for the monoculture kenaf. This can be explained by the higher biomass of the kenaf in rotation (Figure 1). On the other hand, root-induced soil acidification and cadmium mobilization in the rhizosphere of *S. plumbizincicola* could also have contributed to the higher HM removals for the R-Kenaf

treatment [42]. Besides, this also indicates that the cultivation of *S. plumbizincicola* might have promoted the growth of the kenaf in rotation. This may be caused by the relatively lower metal concentration in soil (Figure 3), which decreased the suppression effects of metals on the kenaf in rotation.

The TF evaluates the ability of a plant to translocate the HM from the root to the aerial part [33]. The results from this study show that both *S. plumbizincicola* and kenaf have a higher TF for Cd and Zn than for Cu and Pb. The TF values for Cd and Zn were higher than one (except for the Kenaf treatment for Cd), which suggests that *S. plumbizincicola* has a better ability to transport Cd and Zn from the roots to the shoots, and the cultivation of *S. plumbizincicola* might have the potential to promote the translocation of Cd in kenaf. The BCF evaluates metal accumulation efficiency [33], and the higher the BCF values, the higher the remediation pace for the contaminated soil. The results also indicate that both crops performed better in terms of Cd and Zn remediation. Compared with *S. plumbizincicola*, the remediation ability of kenaf is negligible for Cd and Zn. However, both crops are not ideal candidates for Cu and Pb remediation.

By comparing the metal accumulations in the aerial part and roots, the results show that the accumulation of Cd, Cu, Pb, and Zn in the roots amounts to 8.7%, 122%, 37.6%, and 9.6%, respectively, of that in the aerial part for *S. plumbizincicola*. The accumulation of Cd, Cu, Pb, and Zn in the roots amount to 16.6%, 80.4%, 75.0%, and 10.9%, respectively, of that in the aerial part for kenaf. Hence, the role of the underground part in phytoremediation cannot be neglected [33]. When we harvest HM accumulators in the field, the roots should also be taken away to elevate the remediation efficiency to some extent.

Above all, the rotation with kenaf (summer) and *S. plumbizincicola* (winter) can not only make full use of the farmland at a time scale but can also make up the shortfall of the monoculture with either kenaf (high biomass but low metal removal) or *S. plumbizincicola* (low biomass but high metal removal). In this study, the removal of Cd, Cu, Pb, and Zn by the rotation of two crops increased by 7.88%, 126%, 33.5%, and 4.39%, respectively, compared with the *S. plumbizincicola* monoculture. Hence, the rotation with kenaf and *S. plumbizincicola* can remove more metals from the soil and accelerate the phytoremediation pace.

Although remediation efficiency can be greatly improved by a kenaf–*S. plumbizincicola* rotation, a new problem occurred. Results showed that a large amount of Zn would be removed by the rotation, particularly by the *S. plumbizincicola*. As a necessary element for plants [43], continual cultivation of *S. plumbizincicola* might lead to a Zn deficiency in the soil. Hence, if the method put forward in the current study is used for arable land remediation, the concentrations of nutrient elements must be monitored periodically to avoid decreasing soil fertility.

## 5. Conclusions

Through the present study, several conclusions can be drawn:

- (1) Kenaf can survive under combined HM contamination and can still obtain a huge amount of aerial biomass (17–21 Mg/ha) but cannot accumulate much metal in one season.
- (2) *S. plumbizincicola* can be used for combined HM contamination remediation for its excellent removal ability but its aerial biomass is quite low (7.5–7.9 Mg/ha).
- (3) Rotation with kenaf and *S. plumbizincicola* can greatly improve the remediation efficiency (HM removal for Cd was 7.88%, for Cu 126%, for Pb 33.5%, and for Zn 4.39% higher than *S. plumbizincicola* monoculture) and supply a huge amount of industrial raw material at the same time.
- (4) The underground part of HM-accumulators also plays an important role in phytoremediation and the roots should also be harvested to elevate the remediation efficiency.
- (5) Rotation with bast fiber crops and hyperaccumulators can accelerate the pace of phytoremediation and produce economic value by reusing contaminated farmlands.

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and editing, Y.G., X.C., C.Q., H.Q. and X.Z.; visualization, S.G. and X.Z.; supervision, X.Z.; project administration, S.G. and X.Z.; funding acquisition, X.Z. All authors have read and agreed to the published version of the manuscript.

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