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Chemically Catalyzed Phytoextraction for Sustainable Cleanup of Soil Lead Contamination in a Community Garden in Jersey City, New Jersey

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Abstract: Soil lead (Pb) contamination in Pb paint-contaminated homes is a serious health risk in urban areas. Phytoextraction is a green and sustainable technology for soil Pb remediation, but its efficiency depends on the geochemical partitioning of Pb in soil. Following successful laboratory, greenhouse, and panel experiments, a field study was conducted to demonstrate the effectiveness of a chemically catalyzed phytoextraction model for Pb removal. A biodegradable chelating agent, ethylenediaminedisuccinic acid (EDDS) was applied during Pb phytoextraction by vetiver grass (*Chrysopogon zizanioides*) in a Pb-contaminated community garden in Jersey City, New Jersey. Results showed that soil Pb concentration was reduced from 1144 to 359 mg/kg in 3 years, despite ongoing Pb input to the field plots from a nearby construction site. EDDS was effective in converting non-plant-available forms of Pb (i.e., carbonate-bound, oxide-bound, and organic-bound forms) to plant-available forms (i.e., water-soluble and exchangeable forms). With EDDS application, vetiver roots accumulated 532, 231, and 401 mg/kg of Pb in Years 1, 2, and 3, respectively, which were higher than the values obtained without EDDS applications (228, 154, and 214 mg/kg). This field study demonstrated the effectiveness of a chemically catalyzed phytoextraction model for Pb removal from urban soils.

Keywords: soil lead contamination; lead phytoextraction; lead-based paint; biodegradable chelating agent; vetiver grass (*Chrysopogon zizanioides*)

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1. Introduction

Soil lead (Pb) contamination is a significant environmental and health concern in urban areas [1,2]. Two main sources of Pb contamination in urban locations are leaded gasoline and Pb-based paints [3]. For residential areas, a major soil Pb contributor is deteriorating Pb-based paints in homes built before 1978, when the sale of Pb-based paint for residential properties was banned [4]. However, a significant number of houses in the U.S. still contain Pb-based paint. The deteriorating Pb-based paint from both inside and outside the house could become accessible to humans, particularly children, when paint chips or dust is mixed with soil. The dust generated from the deterioration of Pb-based paint and construction activities can also accumulate on the rooftops of residential buildings [5], which is an additional source of Pb contamination in urban areas. High levels of Pb and other pollutants have been identified in the rooftop runoff [6,7]. The Pb concentrations in urban residential soil are highly correlated with blood lead levels in children [8,9]. Several studies have shown that remediation of yard soil in Pb paint-contaminated homes results in a substantial lowering of blood Pb levels in children [9]. Similar to residential backyards, community gardens located near residential properties are also contaminated by the deterioration of Pb-based paints. Soil Pb in community gardens poses a risk to human health both directly and indirectly. Residents can be exposed to Pb directly from garden soil or dust and indirectly via the consumption of vegetables and fruits grown in these gardens that may have accumulated Pb. Thus, there is a need to remediate Pb-contaminated soil in community gardens, which are popular for growing vegetables in urban areas.

A variety of approaches have been explored for soil Pb remediation, including physical remediation (e.g., excavation, replacement, and capping), chemical remediation (e.g., soil washing and adding stabilizing agents), and biological remediation (e.g., phytoremediation and microbial treatment) [10,11]. Among all these remediation methods, phytoremediation, particularly phytoextraction, is an environment-friendly, effective, and sustainable way to remove Pb from soil [12]. Phytoextraction is defined as the use of green plants to extract Pb from contaminated soils [13–15]. Previous studies have identified many plant species that can accumulate heavy metals, including Pb, but the total removal of contaminants depends on a variety of factors, including soil properties, Pb concentrations and geochemical speciation in soil, Pb uptake in the plant tissue, and the biomass of the plant [16]. Fast-growing plants that can accumulate Pb at high concentrations are preferred in phytoextraction processes. The overall efficiency of phytoextraction heavily depends on the existing forms of Pb in soil, since only the plant-available forms of Pb can be phytoextracted. Chelating agents have been applied to enhance phytoextraction by converting the non-plant-available Pb forms to plant-available forms for enhanced plant uptake [17].

Our previous studies indicated that a biodegradable chelating agent, ethylenediaminedisuccinic acid (EDDS), in combination with a Pb-tolerant perennial grass, vetiver (*Chrysopogon zizanioides*), was able to significantly enhance Pb phytoextraction in laboratory, greenhouse, and panel studies [18,19]. Vetiver grass is a fast-growing, non-invasive, and high-biomass grass that can tolerate heavy metals and extreme climatic variations such as drought, flood, and extreme temperatures [20]. The main goal of this study was to demonstrate the effectiveness of this chemically catalyzed phytoextraction model in a field-scale study. Vetiver grass was used for the removal of soil Pb in a community garden in Jersey City, New Jersey. EDDS was applied to the Pb-contaminated soil to assist Pb phytoextraction by vetiver grass.

2. Materials and Methods

2.1. Experimental Design

A field study was conducted in a community garden for approximately 3 years (June 2020 to February 2023) in Jersey City, New Jersey. The community garden was selected as the study site because of its Pb contamination. Initial soil Pb screening was performed using a portable X-ray fluorescence spectrometer. Three plots were set up in the field: a vetiver treatment plot, a vetiver control plot, and a bermudagrass control plot. The vetiver treatment plot (1.8 m $\times$ 1.8 m, Figure S1 in supplemental material) was tilled and planted with vetiver grass at an interval of 15 cm. Vetiver grass slips were purchased from Mosquito Hawk Farms LLC, Anahuac, TX, USA, and initially cultivated in the greenhouse for 2 months before being transferred to the field. There were 144 vetiver plants in the vetiver treatment plot. Two lysimeters to collect leachate samples and one BSNE (Big Spring Number Eight, Custom Products, Midland, TX, USA) dust sampler to collect dust samples were set in the vetiver treatment plot.

The bermudagrass control plot was identical to the vetiver treatment plot, except that bermudagrass (seeds purchased from Home Depot, Newark, NJ, USA) was sown instead of vetiver grass. The vetiver control plot (0.6 m $\times$ 0.6 m, Figure S1) was in a smaller-sized plot with one lysimeter and one BSNE dust sampler installed. EDDS solution was evenly sprayed on the vetiver treatment plot only, while no EDDS was applied to the other two control plots. Two cycles of EDDS applications (40 L each cycle at a concentration of 2 mmol/L in Year 1 and 10 mmol/L in Years 2 and 3) were performed each year on the vetiver treatment plot. The increase in EDDS dosage was made because unexpectedly, a construction project was initiated next to our plot, in which renovation of a Pb paint-contaminated building was carried out. The project started during late Year 1 and continued during Year 2. Excessive Pb was introduced from this construction site into our plot through dust and stormwater runoff. In Year 3, a hydrological barrier was placed between our field plot and the construction debris to minimize Pb input into our plots by stormwater flow (Figure S2). Soil, leachate, plant, and dust samples were collected before and 3 weeks after each EDDS application. Vetiver grass was trimmed to 1 m in height and left to grow for another 6 weeks before the second round of EDDS treatment and sampling were performed.

2.2. Sample Treatment and Analysis

Soil samples from 9 evenly distributed spots were collected from each plot and mixed to make a composite sample. Soil characteristics of the community garden soil were analyzed using the soil sample before planting. The collected composite soil samples were first air-dried and then sieved (2 mm) before being assayed for pH, electric conductivity (EC), and organic matter content using methods described by Attinti et al. [19]. Total Pb, Fe, and Al concentrations in soil were analyzed using inductively coupled plasma optical emission spectroscopy (ICP-OES, Agilent Technologies 5100, Agilent Technologies, Santa Clara, CA, USA) after acid digestion following Method 3050B developed by U.S. Environmental Protection Agency (USEPA) [21]. After planting, soil, dust, and leachate samples were taken before and after EDDS treatments for Pb analysis. The Pb concentrations in the collected soil, dust, and leachate samples were also analyzed by ICP-OES after acid digestion, which was similar to the composite soil samples. Simultaneously, plant samples were taken, washed, and air-dried before roots and shoots were assayed separately for Pb concentrations. To identify the impact of EDDS on the existing forms of Pb in soil, six geochemical fractions of soil Pb (i.e., F1 water-soluble, F2 exchangeable, F3 carbonate-bound, F4 oxide-bound, F5 organic-bound, and F6 residual silicate-bound) were analyzed as described by Tessier et al. [22] with a few modifications as described by Zhang et al. [18]. All sampling and analyses were performed in triplicate.

2.3. Data Analysis

Data were statistically analyzed using the JMP statistical software package (JMP 14, SAS Institute Inc., Cary, NC, USA). Tukey–Kramer honestly significant difference (HSD) test ($\alpha = 0.05$) was performed to determine the significant differences among different treatment means.

3. Results and Discussion

3.1. Physicochemical Properties of Community Garden Soil

The physicochemical properties of the community garden soil were characterized at the beginning of this field study (Table S1). Results show that the community garden soil was slightly acidic with an average pH of 6.04, which was consistent with previous literature. Most soils in New Jersey are naturally acidic. Hagmann et al. (2015) reported soil pH values ranged from 4.85 to 6.10 in their urban brownfield site in Jersey City, New Jersey [23]. The pH values of the backyard soil of residential properties in our previous study in Jersey City, New Jersey, ranged from 5.32 to 6.14 [18]. The electrical conductivity of the soil was 317 µS/cm, which was higher than that of the Baltimore residential soils (179–242 µS/cm) but lower than those of the two San Antonio residential soils (401 and 428 µS/cm) used in our previous Pb phytoextraction study [19]. The organic matter content (13.4%) of the soil from the community garden was much higher than those from both Baltimore and San Antonio residential soils (0.8–2.4%), which can be explained by the residents adding compost or other soil amendments to grow flowers, vegetables, and fruits in the community garden. The average Pb concentration in the composite soil sample was 906 mg/kg, and the concentrations of two major metals, Al and Fe, were 9611 and 13,517 mg/kg, respectively. Our previous studies have shown that plant availability of soil Pb depends on several soil properties, the most important being soil pH, soil organic matter content, and iron and aluminum oxide/hydroxide content. Pb exhibits increased mobility at acidic pH. High soil organic matter and high levels of iron and aluminum oxides immobilize Pb by forming complexes that decrease Pb mobility in soil [24].

3.2. Lead Concentrations in Soil, Leachate, and Dust Samples

Soil samples from all three plots in the community garden were collected and analyzed for total Pb concentrations at four time points during each year: before first EDDS application, after first EDDS application, before second EDDS application, and after second EDDS application. For the main plot (i.e., vetiver treatment plot), the total Pb concentration in the soil showed a significant decrease from 1144 to 573 mg/kg after the first round of EDDS application in Year 1 (Figure 1a). In comparison, there were no significant changes for either the vetiver control plot or the bermudagrass control plot where EDDS was absent. Previous literature indicated that chelating agents facilitated the formation of strong metal–ligand coordination compounds that are more favored for plant uptake [25]. The chelation of metals is essential in increasing the solubility of Pb and the following remediation of Pb-contaminated soil through phytoextraction [24,26]. Previous studies also demonstrated the efficacy of EDDS in solubilizing non-plant-available Pb to plant-available forms [9,10], and the presence of complexes containing Pb and the chelating agent was identified in vetiver plant tissues [27], which proved the function of EDDS in promoting Pb uptake by vetiver grass. Vetiver grass can accumulate up to 19,800 mg/kg Pb in its root tissue [28]. During the second round of EDDS application in Year 1, construction and renovation activities started in the adjacent building that was approximately 4 feet away from our plots. The active weathering of exterior Pb-based paint and construction activities brought extra Pb to the field through dust and stormwater flow (Figure S2) [29,30], which contributed to the increased soil Pb concentrations in both control plots (Figure 1b,c). Although soil Pb concentration dropped during the second round of EDDS application, the total Pb concentration after the second EDDS application was similar to that after the first EDDS application due to continuous Pb input to the plot (Figure 1a). The building construction and renovation activities continued in Year 2, leading to insignificant changes in Pb concentrations for all plots. In Year 3 when there were no construction activities, and an additional hydrological barrier was placed to protect our plots, a 42.6% decrease in Pb concentration was observed after two cycles of EDDS application in the vetiver treatment plot. The final average Pb concentration was 359 mg/kg, which was lower than the USEPA Pb hazard cutoff value of 400 mg/kg for soil in children's play areas. The Pb concentration in the vetiver control plot also dropped from 857 to 589 mg/kg (Figure 1b), which was much slower than the vetiver treatment plot where EDDS was utilized. Although the average soil Pb concentration decreased in the bermudagrass control plot, there were no significant differences among the soils collected during the four sampling periods collected during Year 3 (Figure 1c). The difference in Pb removal between these two control plots could be explained by the Pb accumulation capacities of vetiver grass and bermudagrass. While vetiver grass was able to accumulate Pb at 19,800 mg/kg in the root and 3350 mg/kg in the shoot, bermudagrass showed much lower accumulation of Pb at concentrations of 1000–2000 mg/kg [28,31].

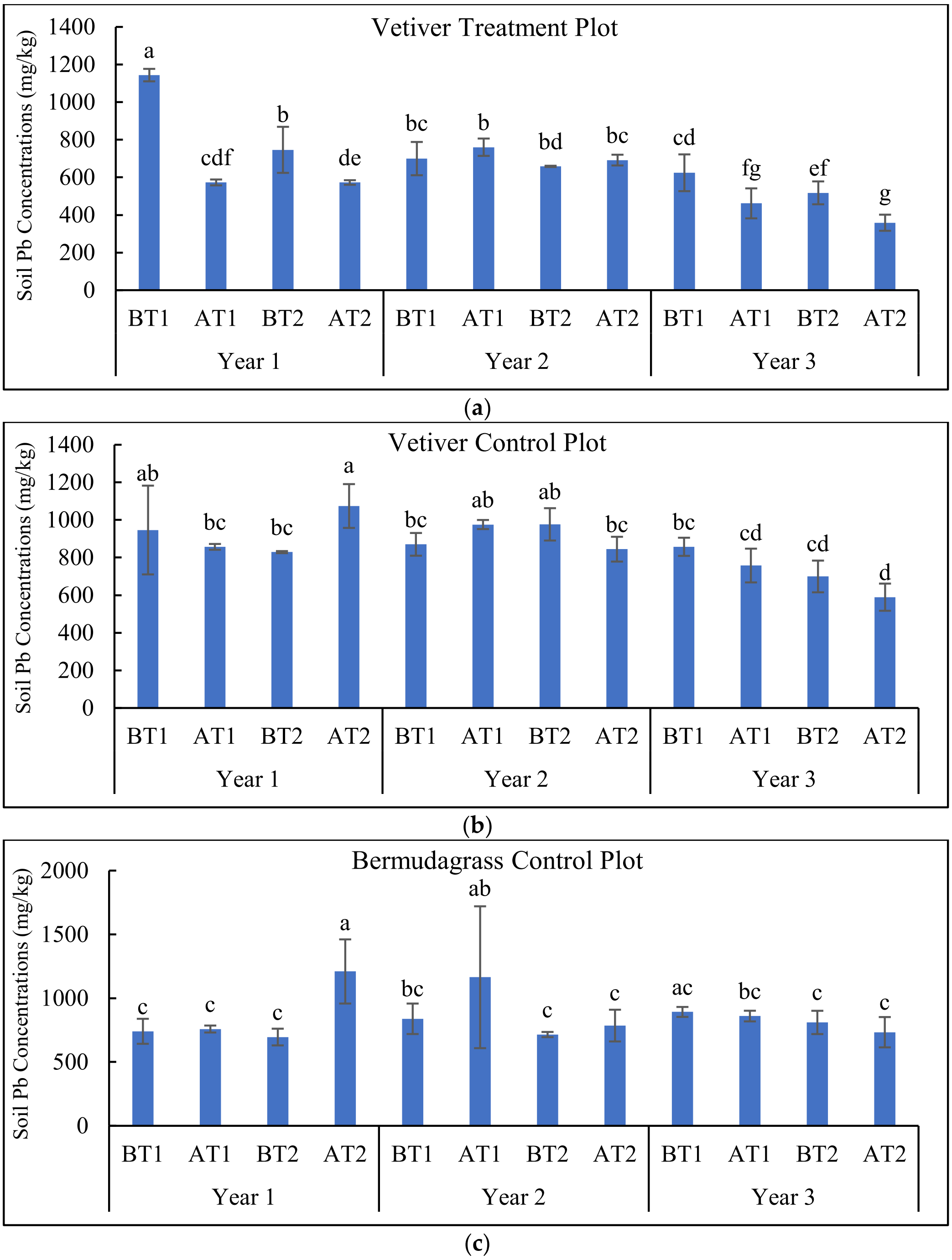


Figure 1. Soil Pb concentrations (mg/kg) in plots with different treatments: (**a**) vetiver treatment plot; (**b**) vetiver control plot; (**c**) bermudagrass control plot. BT1 = before 1st EDDS treatment, AT1 = after 1st EDDS treatment, BT2 = before 2nd EDDS treatment, AT2 = after 2nd EDDS treatment. Data are shown as mean (n = 3) ± standard deviation. Different letters in the same plot correspond to statistically significant differences ($p < 0.05$).

The average Pb accumulations in dust and Pb concentrations in leachate samples each year during all 3 years are shown in Table 1. As expected, the dust Pb accumulations during the 4-month period in Year 2 were higher than the 4-month accumulation in any other year due to construction activities, which also led to increased soil Pb concentrations (Figure 1). The active weathering of paint chips (Figure S2) contributed to the Pb accumulation in both dust and soil, as the Pb concentration in weathered paint chips was measured at 1703 mg/kg. The leachate Pb concentrations were all below 1 mg/L, regardless of the difference in plant species or EDDS applications. After applying EDDS at 10 mmol/L in Years 2 and 3, the Pb concentrations in leachate for the vetiver treatment plot were slightly higher than those in the vetiver control plot, which may imply the mobilization of Pb from non-soluble forms to soluble forms by EDDS [24,32,33]. The low leachate Pb concentration in the vetiver treatment plot also indicated that vetiver grass was able to uptake mobilized Pb instead of excessive Pb infiltrating into the ground and contaminating groundwater.

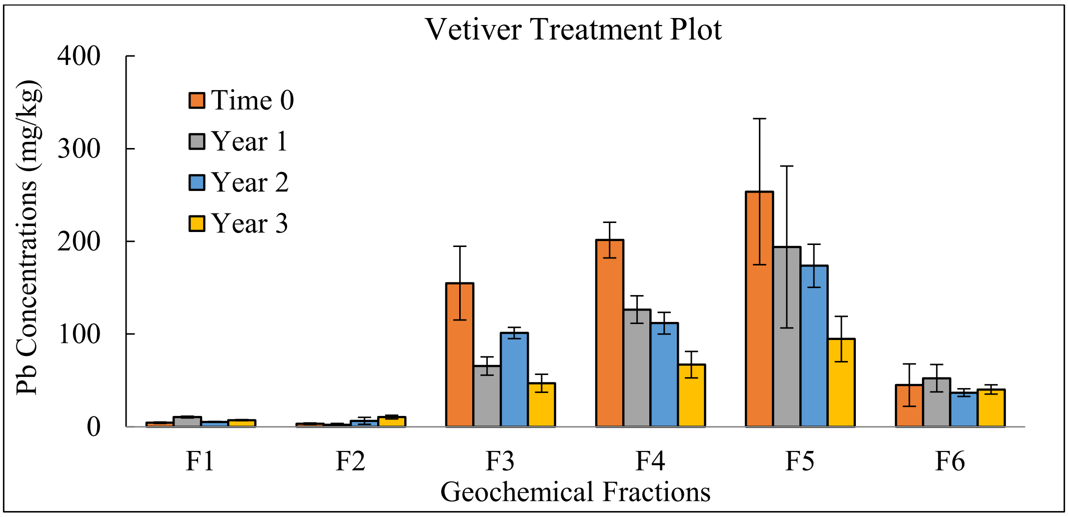
Table 1. Dust Pb accumulation (mg) and leachate Pb concentrations (mg/L) in the plots.

Time and Sample Types		Vetiver Treatment Plot	Vetiver Control'Plot	Bermudagrass Control Plot
Year 1	Dust	0.32	0.46	0.54
	Leachate	0.31	0.38	0.37
Year 2	Dust	0.80	0.51	0.49
	Leachate	0.73	0.66	0.80
Year 3	Dust	0.35	0.41	0.39
	Leachate	0.83	0.71	0.75

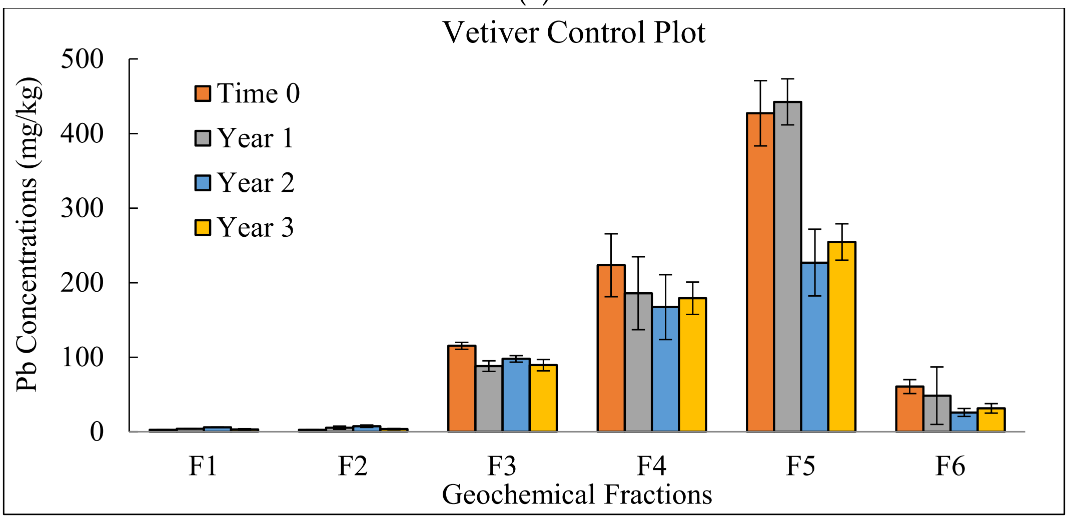
Note: Pb was accumulated in dust during a 4-month period; Pb concentration for each plot was shown as the average values during 4 sampling events (i.e., before 1st EDDS, after 1st EDDS, before 2nd EDDS, and after 2nd EDDS) each year.

3.3. Effect of EDDS on Geochemical Fractions of Soil Lead

The impact of EDDS on soil Pb forms was explored by analyzing soil Pb geochemical fractions. Figure 2a shows the distribution of Pb in six geochemical fractions in the vetiver treatment plot at the beginning of the field study (Time 0) and after the second EDDS treatment in each year. For all three plots, the highest Pb geochemical fraction is F5 organic-bound Pb, followed by F4 oxide-bound Pb (Figure 2). The abundance of organic-bound Pb was due to the high organic content (13.40%) in the community garden soil, compared to other soils in residential properties [18]. The high concentrations of metals, mainly Al (9611 mg/kg) and Fe (13,517 mg/kg), may contribute to the oxide-bound Pb in the community garden soil, which was consistent with our previous study in Jersey City, New Jersey [18]. There was a general trend of concentrations of carbonate-bound Pb (F3), oxide-bound Pb (F4), and organic-bound Pb (F5) decreasing during the experimental period. The Pb concentrations for all these geochemical fractions after the second EDDS application in Year 3 were significantly different from those at the beginning of the study. Specifically, the Pb concentrations in F3, F4, and F5 fractions dropped from 155, 201, and 254 to 47, 67, and 95 mg/kg, respectively (Figure 2a). These Pb geochemical fractions were previously reported to be converted into water-soluble (F1) and exchangeable (F2) fractions because of the use of chelating agents for Pb mobilization [18,34–36]. Research by Attinti et al. [19] indicated an increase in the F5 fraction for Baltimore soils, and Zhang et al. [18] showed an increase in the F4 fraction but a decrease in the F5 fraction for Jersey City and San Antonio soils after the addition of EDDS as a chelating agent. The residual silicate-bound Pb (F6) did not change significantly in this study, as it is the most difficult geochemical fraction to mobilize. However, Li et al. [34] showed a decrease in the F6 fraction using EDTA as the chelating agent, and Zhang et al. [18] demonstrated the mobilization of F6 by >50% in a laboratory study using EDDS. Those discrepancies could be caused by using different types of chelating agents or by other conditions, such as pH and the presence of other ions [26,37]. In comparison, the Pb geochemical fractions generally remained unchanged for both control plots, which further proved the function of EDDS in converting non-plant-available forms of Pb (F3–F6) to plant-available forms (F1 and F2) to facilitate phytoextraction.



(**a**)



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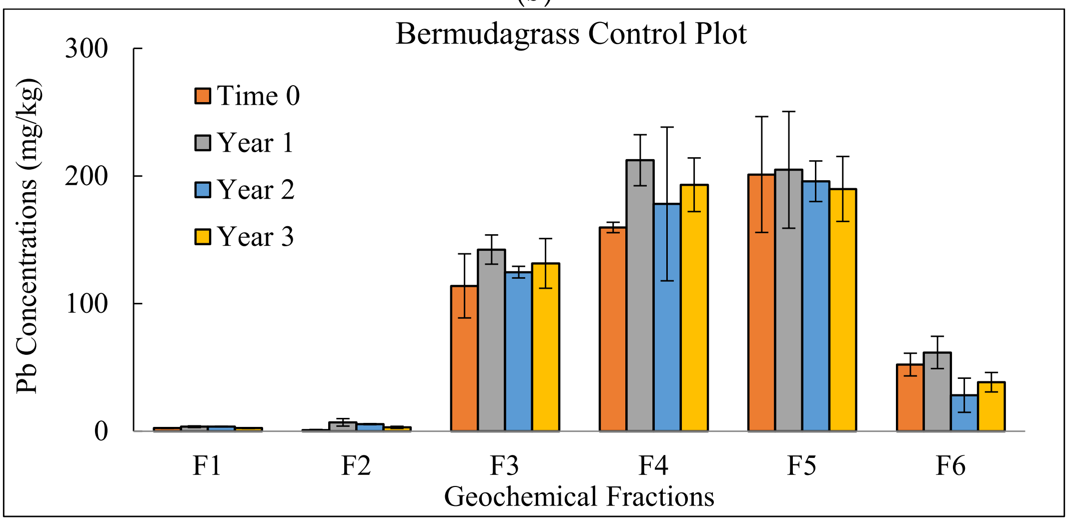

(**c**)

Figure 2. Geochemical fractions of Pb in soils after two EDDS treatments each year: (**a**) vetiver treatment plot; (**b**) vetiver control plot; (**c**) bermudagrass control plot. Data are shown as mean (n = 3) ± standard deviation.

3.4. Lead Uptake by Vetiver and Bermudagrass

The Pb uptake by both vetiver and bermudagrass was quantified. Lead concentrations in the roots and shoots are shown in Figure 3. There was a large accumulation of Pb in the root of vetiver grass each year (Figure 3a–c). Mean Pb concentration increased by 319%, 124%, and 178% in vetiver root for Years 1, 2, and 3, respectively, in the vetiver treatment plot. The corresponding increases for the vetiver control plot were 49%, 144%, and 118%, respectively. The Pb concentrations for vetiver root in both vetiver plots were similar at the beginning of each year, but the concentrations after EDDS applications (532, 231, and 401 mg/kg in Years 1, 2, and 3, respectively) were much higher than those without EDDS (228, 154, and 214 mg/kg in Years 1, 2, and 3, respectively). During the field study, vetiver grass did not show any phytotoxic symptoms after taking up Pb from the soil (Figure S3). The results are consistent with our previous panel study showing that EDDS promoted the uptake of Pb by vetiver grass [19]. Lead accumulation in the shoot of vetiver grass grown in the vetiver treatment plot was also more than that in the vetiver control plot (Figure 3d–f), indicating that more Pb was translocated from vetiver root to shoot in the presence of EDDS. Several studies have shown that chelating agents enhance both metal availability and its translocation from plant root to shoot [38,39]. The results shown in Figure 3 are consistent with previous literature regarding enhanced plant uptake and translocation of Pb with the assistance of chelating agents, such as EDTA, citric acid, and EDDS [40–42].

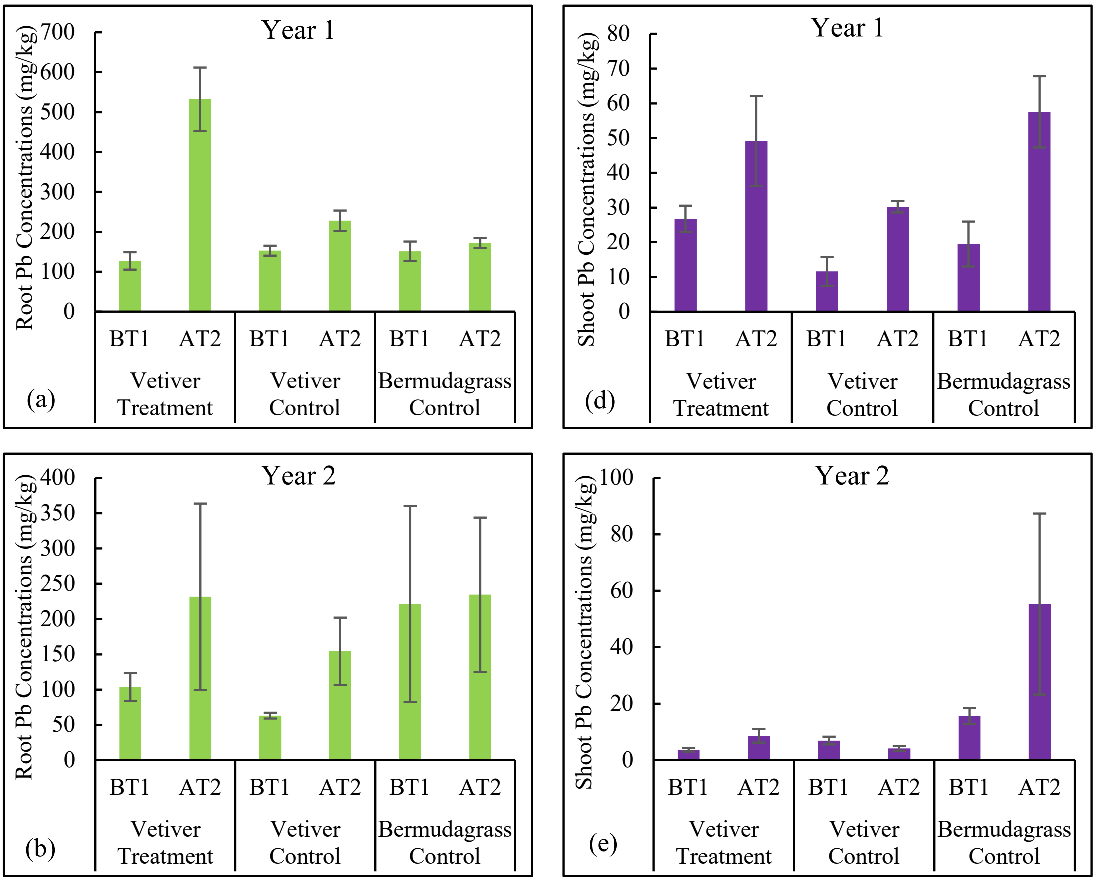

Figure 3. *Cont.*

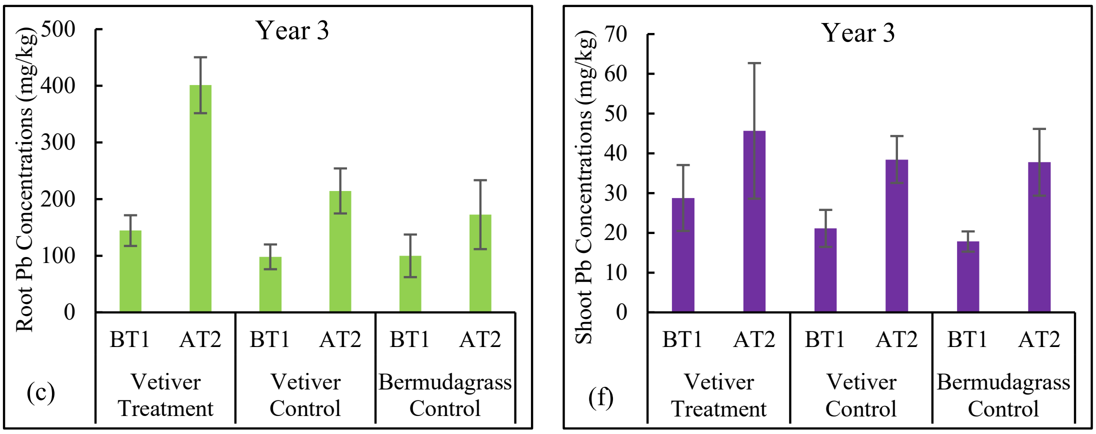

Figure 3. Pb uptake by tissues (root: (**a**–**c**); shoot: (**d**–**f**)) of vetiver grass and bermudagrass grown in Pb-contaminated community garden soil. BT1 = before 1st EDDS treatment, AT2 = after 2nd EDDS treatment. Data are shown as mean (n = 3) ± standard deviation.

The Pb accumulation in roots of vetiver and bermudagrass at the end of each year for both control plots did not show significant differences (Figure 3a–c). However, higher Pb concentrations were found in the shoots of bermudagrass during Years 1 and 2. These results indicate that vetiver grass had lower Pb translocation factors (0.13, 0.03, and 0.18 in Years 1, 2, and 3, respectively) than bermudagrass (0.33, 0.24, and 0.22 in Years 1, 2, and 3, respectively). Although Pb uptake by bermudagrass was slightly higher in terms of total Pb concentrations per dry biomass weight, more Pb was removed by vetiver (Figure 1) due to its massive root and shoot systems with much higher biomass. Bermudagrass was also previously reported to extract less Pb and Ni from contaminated soils than other common grasses such as fescue grass [43]. This study also showed that EDDS is an effective chelating agent for Pb phytoextraction and can be used instead of typical non-biodegradable chelating agents such as EDTA that are toxic to plants [44].

4. Conclusions

This study employed an EDDS-catalyzed phytoextraction method for Pb removal from the soil in a community garden using vetiver grass. Results showed that the initial Pb concentration of 1144 mg/kg was reduced to 359 mg/kg in 3 years. The field site experienced continuous input of Pb from an adjacent construction site that actively released Pb-based paint, which impacted the soil Pb concentration. We believe that without this continuous input of Pb, the phytoextraction process would have been faster. The efficacy of EDDS in converting non-plant-available Pb to plant-available forms was demonstrated by tracking the geochemical forms of Pb throughout the study. Vetiver grass accumulated large amounts of Pb in its roots without showing any phytotoxicity. The results show that the chemically catalyzed phytoextraction model we developed is an effective and sustainable method for addressing Pb contamination in urban soils. This study was limited to a community garden in New Jersey with acidic soil. It would be important to conduct these studies on soils of varying pH and organic matter content to understand the effectiveness of EDDS. In addition, vetiver is a tropical/sub-tropical grass, which limits its use in colder climates.

This field demonstration study was based on our previous investigation of this EDDS-catalyzed Pb phytoextraction model in laboratory- and pilot-scale experiments. The successful removal of Pb with a final Pb concentration at <400 mg/kg indicated that this model could be practiced in urban areas, such as community gardens and residential properties, that have high Pb concentrations. Since vetiver grass is a fast-growing perennial grass that can tolerate extreme climatic variations (e.g., drought, flood, and extreme temperatures), less maintenance would be required compared to other plants used for phytoextraction, making it a viable solution for broad application in urban areas. The experimental protocol in this field study was easy to perform, involving regular watering and weeding, which could be performed by community garden users or homeowners with minimal instructions. In addition, the collected vetiver grass after extracting Pb from residential or community garden soils can be further utilized to produce second-generation bioethanol, which we have shown recently [45]. Thus, a circular economy model could be feasible and worth investigating in the future.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su15097492/s1, Figure S1: Plot setting for the field study experiments; Figure S2: Overall picture of the field site; Figure S3: Vetiver grass growth in the vetiver treatment plot in the community garden; Table S1: Properties of the composite soil samples in the community garden.

Author Contributions: D.S. and R.D. conceptualized and designed the study and obtained the funding. Z.Z. and F.L. performed field studies and sample analysis. Z.Z. wrote the first draft of the paper. R.D. and D.S. reviewed and edited the paper. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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