sustainability

MDPI

Article

Electrokinetic-Assisted Phytoremediation of Pb-Contaminated Soil: Influences of Periodic Polarity Reversal Direct Current Field

Hayishaer Mulati [1], Anwar Mamat [1,*], Nuerla Ailijiang [2], Lu Jiang [1], Ning Li [1], Yuanfang Hu [1] and Yuhong Su [1,*]

1 College of Chemical Engineering, Xinjiang University, Urumqi 830017, China
2 College of Ecology and Environment, Xinjiang University, Urumqi 830017, China
* Correspondence: anwarm@xju.edu.cn (A.M.); suyuhong@xju.edu.cn (Y.S.)

Abstract: The effect of electric stimulation on the phytoremediation of Pb-contaminated soil by tall fescue and wheat seedlings was investigated by monitoring the time-dependent changes in soil properties and Pb accumulation in plants with or without a direct current (DC) exchange field. The results showed that plants could strengthen electrical conductivity (EC) and current in the soil environment. The periodic exchange electrode helped maintain a stable soil pH. Electric stimulation enhanced phytoremediation efficiency, and the enhancement effect increased with plant growth. Compared with the treatments without a DC exchange electric field, the Pb content in plots with tall fescue and wheat seedlings under DC exchange electric field increased 107–250.6% and 32.7–84.4%, respectively, after 30 d of planting. DC exchange electric field significantly increased the Pb enrichment coefficient of tall fescue and wheat seedlings. The upward transport of Pb from wheat seedling roots to shoots was greatly promoted by electric stimulation for 18 d. However, Pb transport in both plants was restrained after 18 d of electric stimulation. Additionally, DC exchange electric stimulation can improve the phytoremediation of heavy-metal-contaminated soil. More attention should be paid to the selection and testing of promising hyperaccumulators for electrokinetic-assisted phytoremediation of Pb-contaminated soil.

Keywords: direct current exchange electric field; phytoremediation; heavy metals; Pb; soil section

Citation: Mulati, H.; Mamat, A.; Ailijiang, N.; Jiang, L.; Li, N.; Hu, Y.; Su, Y. Electrokinetic-Assisted Phytoremediation of Pb-Contaminated Soil: Influences of Periodic Polarity Reversal Direct Current Field. *Sustainability* **2023**, *15*, 8439. https://doi.org/10.3390/su15118439

Academic Editors: Robson Andreazza and FILIPE Selau Carlos

Received: 29 March 2023
Revised: 17 May 2023
Accepted: 21 May 2023
Published: 23 May 2023

1. Introduction

Exposure range and contact frequency of human beings with heavy metals is increasing with urbanization and industrialization [1]. The treatment of heavy metal pollution is a global problem because it threatens the safety of the soil environment and food production. Lead (Pb) is an important soil pollutant owing to its high toxicity, non-degradability, and bioavailability [2]. Pb in soil has been mainly sourced from transport emissions (leaded gasoline), mining, smelting, and e-waste over the past three decades (1990–2017) [3]. It is well known that Pb can accumulate in crops grown in contaminated soil and enter the human body via food-chain transmission [4]. It poses serious health risks to humans and animals, even at very low concentrations in the environment [5]. According to a national survey of soil pollution in China, the excess rate of Pb accumulation was 1.5%, and the average concentration was 1.35 × 103 mg kg^{-1} [6] in the city soil. To mitigate the ecological risks, 90% of the heavy-metal-polluted soil in China were planned to be purified by 2030 [7]. Traditional remediation technologies for heavy-metal contamination of soil include chemical, biological, and physical methods. Physical and chemical remediation technologies, such as washing, immobilization and stabilization, and osmotic reaction walls, are widely used because they are mature and can quickly and effectively isolate heavy metals from soil [8]. However, physical methods have the shortcomings of large-scale engineering, high transport and treatment costs, and a major impact on soil properties and function [9]. Chemical remediation technologies can also destroy the soil matrix structure and physicochemical properties, causing nutrient loss [10]. Moreover, chemical reagents remain for a long time, causing secondary pollution [11]. These shortcomings restrict the use of chemical technologies within a narrow repair scope [12]. As a biological technology, phytoremediation has many advantages, such as low treatment cost, effectiveness for many heavy metal pollutants, and little influence on the environment [13]. There are also some restrictive factors, such as slow growth and low biomass of hyperaccumulators, long remediation cycles, low bioavailability of heavy metals, and difficult remediation of deep soil pollution, which restrict their practical application [14]. At present, it is still necessary to develop efficient and low-cost remediation technologies for removing heavy metals from soil in larger areas [15]. Novel technologies combining multiple methods may be the correct development direction to compensate for the limitations of typical remediation technologies.

Compared with other methods, electrokinetic remediation has many advantages, such as high efficiency, simple and economical operation, and non-destruction of the natural ecological environment, which receives domestic and foreign scholars' attention [16]. Electrodynamics can effectively improve the soil pollutant absorption and enrichment potential of plants [17]. The organic combination of electrodynamics and phytoremediation may compensate for their respective shortcomings and produce synergistic effects, activating heavy metals in the soil and improving bioavailability, thereby enhancing the uptake and transport of heavy metals by plants and improving the efficiency of phytoremediation. An important consideration is that electrodynamics may affect plant growth by changing the soil pH (especially in the anode region) and causing heavy metal stress. It has been reported that the growth of many plants (ryegrass, potato, Indian mustard, etc.) has not been adversely affected; moreover, the biomass of some plants has increased significantly [18]. The external electric field promotes plant growth partly because of the remediation effect of plants on the heavy metals (Cu, Pb, Zn, and Cd) in soil [19]. The enrichment ability of Indian mustard in high Pb-contaminated soil under the condition of electrodynamics and ethylenediamine tetraacetic acid (EDTA) addition is 2–4 times that of EDTA alone [20]. However, the role of electrodynamics will lead to changes in the physico-chemical properties of soil media, for instance, soil acidity and alkalinity, zeta potential, organic matter, and nutrient activity, which will significantly affect the spatial distribution, morphology, and biological activity of heavy metals. When the direct current (DC) was applied, the soil pH in the cathode area increased significantly, while that in the anode decreased significantly, and the heavy metal content in the soil was spatially redistributed [21]. Electrodynamics can also alter the transport of heavy metals in plants. Lim et al. [20] reported that a DC combined with EDTA promoted the transport of heavy metals from the roots of Indian mustard to the aboveground parts, and enhanced its repair rate and efficiency. Recently, Siyar et al. [22] used vetiver as an enrichment plant to remediate heavy-metal-contaminated soil around a mining area. Studies have shown that the growth of vetiver is inhibited by heavy metals in soil. The application of a DC electric field significantly changed the pH values of the soil, whereas the alternating current (AC) electric field only induced weak changes. The maximum accumulation of metals in vetiver grass occurred at 2DC V cm^{-1}, showing about a 50% enhance in comparison with an AC cell [22]. Plant stabilization, rather than plant extraction, may be the main mechanism on the removal of heavy metals coupled with the DC electric field, whereas the AC electric field has a more prominent influence than DC electric field in promoting the extraction of heavy metals by plants. Li et al. [23] showed that a DC electric field could enhance the plant uptake of different forms of uranium [UO_2, UO_3, $UO_2(NO_3)_2$] by sunflower and Indian mustard, and the difference between the different forms was significant.

The organic combination of electrodynamics and phytoremediation may compensate for their shortcomings and produce synergistic effects, activate soil heavy metals, and improve their bioavailability, thereby improving the uptake, and transport of heavy metals by plants, which may improve phytoremediation efficiency. Crops have the advantages of large biomass, strong stress resistance, and wide adaptability. Compared with hyperaccumulators, different crops have certain advantages in the application of heavy-metal-contaminated soil remediation in farmlands. However, there are few reports on the combination of electrokinetic remediation and phytoremediation for remediating Pb-contaminated soil by the indirect connection between the electrode and soil. The novelty of the work is to explore the law of periodic polarity reversal DC electric field and repair time on the accumulation of Pb in tall fescue and wheat seedlings. Therefore, the main aims of the research were to (i) study the influence of periodic polarity reversal on current, soil pH and electrical conductivity, (ii) explore the effect of DC stimulation on the uptake and accumulation of the Pb in soil by tall fescue and wheat seedlings, and (iii) study the effect of DC stimulation on the upward transport of lead in tall fescue and wheat seedlings.

2. Materials and Methods

2.1. Chemicals

Tall fescue and wheat seeds were procured from the Urumqi Flower Market in Urumqi, China. All chemicals used in this research were analytically pure. The aqueous solutions of all chemicals were prepared with ultrapure water (18.2 MΩ).

2.2. Soil Preparation

The tested soil was gathered from the surface soil (0–20 cm depth) in Bagang Park, Urumqi, Xinjiang, China. After the soil was dried, plant residues and debris were removed. After grinding and passing the soil through a 2-mm mesh sieve, 3 kg of the tested soil was taken into a large, sealed bag, and 800 mL of $Pb(NO_3)_2$ solution with 1025.78 mg L^{-1} concentration was added to the soil (the water content of each kg of soil was approximately 73.7% of the saturated water content of the soil). After stirring, the sealed bag was placed in a cool place for natural spiking for 30 d. After spiking, the soil was dried in a cool place for 5 d. Subsequently, the soil was ground, passed through a 2-mm mesh sieve, and mixed thoroughly. Finally, simulated contaminated soil with a Pb mass concentration of 268.24 ± 5.29 mg kg^{-1} was obtained and stored in sealed bags. The measured physical and chemical properties of the soil are listed in Table 1.

Table 1. Physical and chemical properties of soil.

Test Indicators	pH	Organic Matter Content (g kg^{-1})	Moisture Content (%)	Cation Exchange Capacity (cmol kg^{-1})	Conductivity (ms cm^{-1})	Pb Concentration (mg kg^{-1})
Simulate the soil	7.44 ± 0.03	40.40 ± 0.17	25.7 ± 0.64	16.21 ± 0.46	3.43 ± 0.01	268.24 ± 5.29

2.3. Experiments Setup

This experiment consists of five treatments: a treatment unseeded with DC alone (EK); two treatments without DC, one seeded with tall fescue (FA) and one with wheat (WT); and two treatments with DC, one seeded with tall fescue (FA + EK) and one with wheat (WT + EK). The experimental design is summarized in Table 2.

The electric repair experimental device included a main rectangular plastic mold of 26 cm (length) × 16 cm (width) × 13 cm (height) (1) pasted with two perforated poly (methyl methacrylate) partitions (2). The main body of the device was divided into three parts: the middle part was the soil chamber of 16 cm (length) × 16 cm (width) × 13 cm (height), and the two sides were electrode chambers of 5 cm (length) × 16 cm (width) × 13 cm (height). Filter cloth (500 μm) (3) was pasted on the soil indoor side partition (2). The graphite plate electrode (4) was placed in the electrode chamber. After drilling the top of the graphite plate electrode (4), copper wire (5) was connected to the DC-regulated power supply (6). A multimeter (7) was connected in series between the circuit of the graphite plate electrode (4) and the DC-regulated power supply (6) to monitor the current change in the device. The soil chamber was divided into four sections (S1–S4), from left to right, to plant restoration plants (8). A schematic diagram of the experimental apparatus is shown in Figure 1.

Table 2. Experimental design.

Experiment Number	Processing	Power Mode	Plant Species	Power-Up Time (h)	Repair Time (d)
T1	FA	/	Tall Fescue	0	30
T2	WT	/	Wheat	0	30
T3	EK	Exchange electrodes	/	6	30
T4	FA + EK		Tall Fescue	6	30
T5	FA + EK		Tall Fescue	6	30
T6	FA + EK		Tall Fescue	6	30
T7	WT + EK		Wheat	6	30
T8	WT + EK		Wheat	6	30
T9	WT + EK		Wheat	6	30

Note: EK: a treatment unseeded with DC alone. FA and WT indicated that tall fescue and wheat were sown separately without DC. FA + EK and WT + EK indicated that tall fescue and wheat are sown under DC conditions.

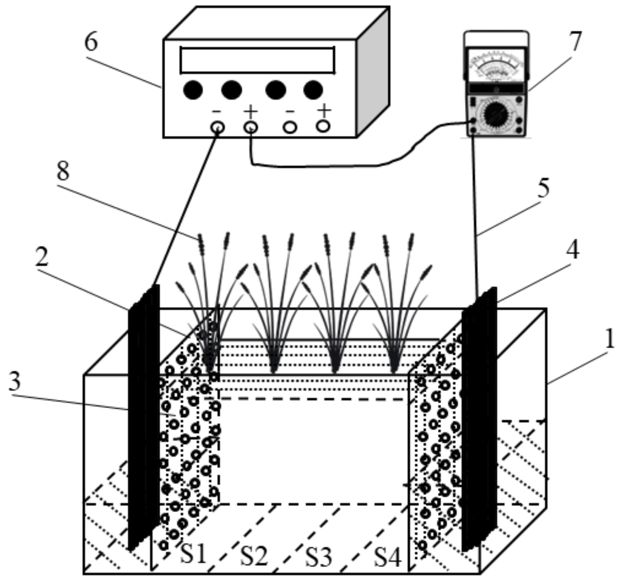

Figure 1. Schematic diagram of the experimental apparatus. Note: 1: Plastic concrete mold; 2: Perforated poly (methyl methacrylate) partitions; 3: filter cloth; 4: graphite plate electrode; 5: copper wire; 6: DC regulated power supply; 7: multimeter; 8: phytoremediation.

Each soil chamber was filled with approximately 3 kg of heavy-metal-contaminated dry soil and sprayed on the soil surface with a spray kettle until it was moist. The soil chamber was divided into four sections, S1–S4, from left to right, to plant tall fescue and wheat seeds, and a layer of soil was evenly spread to allow the seeds to germinate naturally and grow for 20 d. After 20 d of plant growth, 500 mL of tap water was added to the electrode chambers on both sides of the device and allowed to stand for 24 h. After the soil absorbed water evenly, tap water was added again to the liquid level in the electrode chamber at a height of 3 cm; then, tap water was added daily to maintain the electrolyte height at 3 cm. The experiment was carried out in the natural indoor environment of the campus of Xinjiang University in Urumqi, China, and grew for 50 days in the summer of 2022 under the conditions of daytime temperature range of 20–30 °C and nighttime temperature range of 18–25 °C without supplementary lighting. The soil water content was maintained at 60% of the water holding capacity. After 20 d of plant growth, a DC electric field with a constant voltage gradient of 1 V cm^{-1} was applied for 30 d. It was provided for 6 h per day during the daytime, and the electrode polarity was switched daily. The electrode chambers on both sides of the device were placed in the left and right electrode chambers. The current of each device was tested daily at the beginning and end of energization.

2.4. Sample Collection and Extraction

2.4.1. Soil Samples

For the collection of soil samples, the soil chamber was divided into four sections, S1–S4, from the cathode to the anode, and soil samples in the S1–S4 sections were collected separately. The soil samples were air-dried in a cool place for 2–3 d. After grinding, the samples were sieved at 0.150 mm and stored in sealed bags. Soil samples were digested using aqua regia reflux digestion (NY/T 1613-2008). Soil samples (0.3 g; error not more than 0.0003 g) were weighed in a 50 mL digestion tube, and 3 mL of concentrated nitric acid was added to it and incubated for 12 h. Then, the digestion tube was placed in an infrared digestion furnace and heated at 80 °C for 5 h. Then, it was heated to 120 °C for 24 h, then to 130 °C, and heating was continued until 2 mL of the concentrated nitric acid was left. The digestion tube was removed and cooled to 25 °C, and 6 mL of concentrated hydrochloric acid was added to the tube and incubated for 12 h. The digestion tube was then placed in an infrared digestion furnace and heated at 80 °C for 2 h. The temperature was increased to 120 °C and heated continuously until the sample was gray and white. The liquid in the tubes was stored at 25 °C and filtered. The filtered soil sample was diluted to 25 mL in a colorimetric tube for measurement.

2.4.2. Plant Samples

Plant samples in the S1–S4 sections of the soil chamber were collected on days 18 and 30 of energization. The plant roots and shoots were harvested separately, washed with tap water, and rinsed with purified water. Subsequently, roots were soaked in 20 mM disodium ethylenediaminetetraacetic acid (Na_2EDTA) for 15 min to remove metal ions from the surface of the roots, and then rinsed with purified water three times. Plant samples were dried in a 105 °C oven for 30 min, and then dried to constant weight at 70 °C. The samples were ground and passed through a 100-mesh nylon sieve. The sample was placed in a 50 mL digester with 3 mL concentrated nitric acid for 12 h. The digestion tube was then placed in an infrared digestion furnace and heated at 80 °C for 5 h. Then, the temperature was raised to 120 °C for 24 h, and heating was continued until the plant sample disappeared. The liquid in the tube was filtered after it returned to 25 °C. The filtered plant samples were diluted to 10 mL in a colorimetric tube for measurement.

2.5. Analytical Method

2.5.1. Soil Physicochemical Parameters

The soil pH and electrical conductivity (EC) were measured using a pH meter (DELTA 320, Mettler Toledo Instruments Co., Ltd., Shanghai, China) and an EC meter (DDS-310, Shanghai Kangyi Instrument Co., Ltd., Shanghai, China), respectively. The soil sample (5 g) was weighed in a centrifuge tube, and 12.5 mL of carbon dioxide-free water was added to it (soil-water ratio of 1:2.5). Subsequently, the mixture was shaken for 1 h and then kept still for 30 min. The pH meter was calibrated using pH 4.00 (25 °C) and pH 6.86 (25 °C) standard buffer solutions. The probe was immersed in the suspension to directly measure the pH of the sample. After the pH stabilized, the data were recorded.

After the samples were kept still for 30 min, they were centrifuged at 3000 r min^{-1} for 5 min, and then the soil conductivity meter probe was immersed in the supernatant to directly measure and record the readings.

Determination of soil organic matter content used the potassium dichromate volumetric method [24], soil moisture content was determined using the drying weighing method [25], and cation exchange capacity was determined using the barium chloride buffer method [26].

2.5.2. Heavy Metal Content in Soil and Plants

Pb content in the soil and plant samples was determined using flame atomic extraction spectrometry (TAS-990, Beijing General Instrument Co., Ltd., Beijing, China). The determination method was based on the literature review [27].

2.5.3. Parameters of Analysis

Bioaccumulation factor (BCF) and translocation factor (TF) are two parameters widely used in phytoremediation experiments. BCF reflects the ability of plants to extract and enrich heavy metals in soil [28]. It represents the ratio of the heavy metal content in plants to the total amount of heavy metals in the soil. TF is the ratio of heavy metal content in the aboveground part of plants to that in the underground part of plants; it is an important index for evaluating the heavy metal transport capacity of plants [29]. They can be calculated by the following formulas:

$$BCF = \frac{metal\ concentration\ in\ plants}{metal\ concentration\ in\ soil}$$

$$TF = \frac{metal\ concentration\ in\ shoots}{metal\ concentration\ in\ roots}$$

2.6. Quality Control

For quality control, the standard reference samples and reagent blanks were digested and determined simultaneously. Standard reference samples, such as soil samples (GBW07426) and Pb standard solutions (100 mg L^{-1}), were provided by the National Standard Substances Center. The average of calculated recoveries of reference soil samples was 101.3 $\pm$ 3.2%. All samples with tested concentrations were corrected with corresponding recovery rates.

3. Results and Discussion

3.1. Effect of Exchange Electric Field on Pb Uptake by Plants and Pb Content in Soil

At 18 d after electrification (Figure 2a), the content of Pb in the aboveground parts of tall fescue at S2, S3, and S4 of the FA + EK treatment group reduced significantly, by 31.9%, 15.2%, and 33.4%, respectively, whereas the content of Pb in the underground parts of tall fescue at S2, S3, and S4 of the FA + EK treatment group increased significantly, by 18.5%, 15.9%, and 28.2%, respectively, compared with that of the FA treatment group. The content of Pb in the shoots of wheat at S1 and S3 in the WT + EK treatment group increased significantly, by 619.4% and 155.6%, respectively, compared with that of the WT treatment group. After electrification, the content of Pb in the ground parts of wheat at S1 and S2 in the WT + EK treatment group increased significantly, by 22.9% and 49.4%, respectively, compared with that of the WT treatment group. At 30 d, the content of Pb in the aboveground parts of tall fescue at S1, S2, and S3 of the FA + EK treatment group increased significantly, by 102%, 85.3%, and 70.8%, respectively, compared with that of the FA treatment group (Figure 2b). Furthermore, after electrification, the content of Pb in the underground parts of tall fescue at S1, S2, and S3 of the FA + EK treatment group increased significantly, by 250.6%, 107%, and 175.2%, respectively, compared with that of the FA treatment group. At 30 d, compared with the FA + EK and FA treatment groups, the content of Pb in the shoots of wheat at S1–S4 were not significantly different between the WT + EK and WT treatment groups. After electrification, the content of Pb in the underground parts of wheat at S2, S3, and S4 in the WT + EK treatment group increased significantly, by 84.4%, 32.7%, and 55.2%, respectively, compared with that of the WT treatment group.

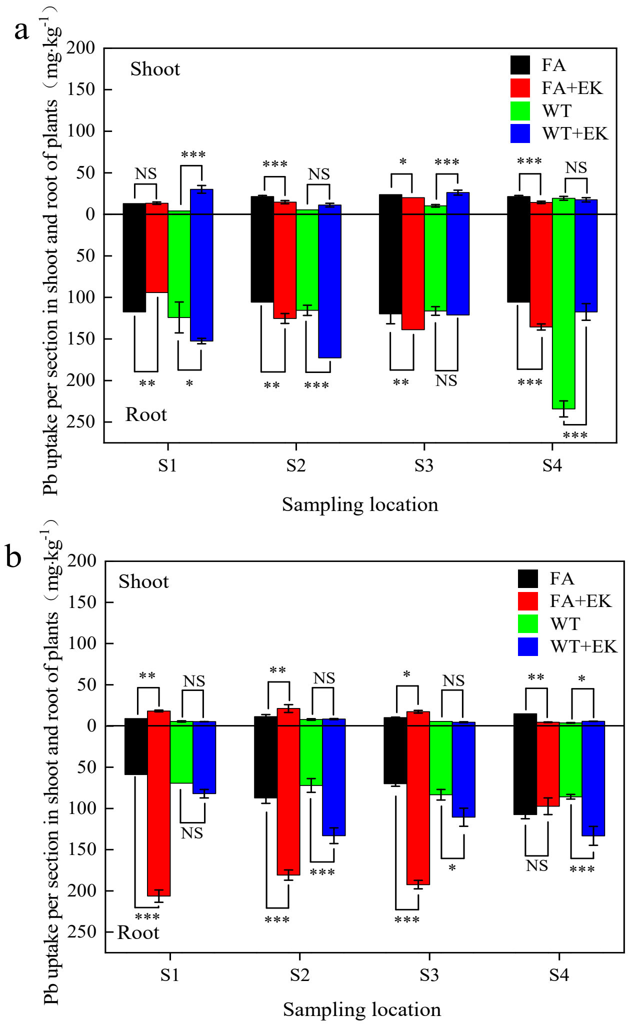

Figure 2. Effects of different treatments on the content of heavy metals in shoot and root segments of plants at different sampling points of 18 days (**a**) and 30 days (**b**). FA and WT indicated that tall fescue and wheat were sown separately without DC. FA + EK and WT + EK indicated that tall fescue and wheat are sown under DC conditions. NS: not significant, *** $p \leq 0.001$; ** $p \leq 0.01$; * $p \leq 0.05$.

The above experimental results show that the exchange electrode could promote the extraction and enrichment of Pb in the soil of tall fescue and wheat. This may be because the application of an electric field can increase the content of soluble heavy metals in the soil and facilitate the extraction and enrichment of heavy metals by plants. Moreover, a DC exchange electric field can significantly enhance the mobility of heavy metals in the soil and promote the directional migration of soluble heavy metals to the roots [30]. The Pb content in the underground parts of tall fescue and wheat seedlings was markedly higher than that in the aboveground part, and Pb was mainly enriched in the roots of plants, which was consistent with the results of Putra et al. [31]. The first step of the plant extraction process was to extract heavy metals into root cells and then transfer heavy metals from the underground part to the aboveground part [31]. Electrification for 30 d promoted the absorption and enrichment of soil Pb in the underground parts of tall fescue and wheat. Electric stimulation improves the efficiency of phytoremediation and increases plant growth. The results showed that the exchange electrode had a certain influence on the Pb content in the aboveground parts of tall fescue and wheat seedlings. The specific performance was found to be stimulated for 18 d, and the current inhibited the upward transport of Pb in tall fescue, whereas it promoted the upward transport of Pb in wheat seedlings. During the 30 d, electrification promoted Pb translocation in tall fescue but did not affect Pb translocation in wheat seedlings. The aboveground parts of plants can be easily harvested and treated (e.g., by burning); thus, the metal content and biomass of the aboveground part of the plants have a great influence on the phytoremediation efficiency. Therefore, the metal content and biomass of the aboveground parts of plants are the most important factors for determining the efficiency of phytoremediation. In general, the impact of an applied electric field on the accumulation of heavy metals in plants varies depending on plant species and soil sections. Electrical field affected phytoremediation efficiency through changed electrochemical and biological processes in different soil profiles, meaning that changes in soil EC may be a significant factor affecting plant growth and metal content. However, the effect of EC changes on plant metal concentration needs further study. The polarity reversal DC electric field keeps the soil pH stable, which is beneficial to the normal growth process of plants. Electrokinetic-assisted phytoremediation was a promising method for Pb-contaminated soil remediation. Specifically, the combination of polarity reversal DC electric field and tall fescue was a good choice for remediation of Pb-contaminated soil.

The change of Pb content in different soil profiles was not significant at 18 days and 30 days (Figure 3). Periodic polarity reversal DC electric field activates Pb in the soil, which causes the increase of the plant root uptake as well as homogenization distribution of Pb in soil. Meanwhile, the reduction of the total Pb content in soil does not show a significant change after phytoremediation due to the low plant biomass.

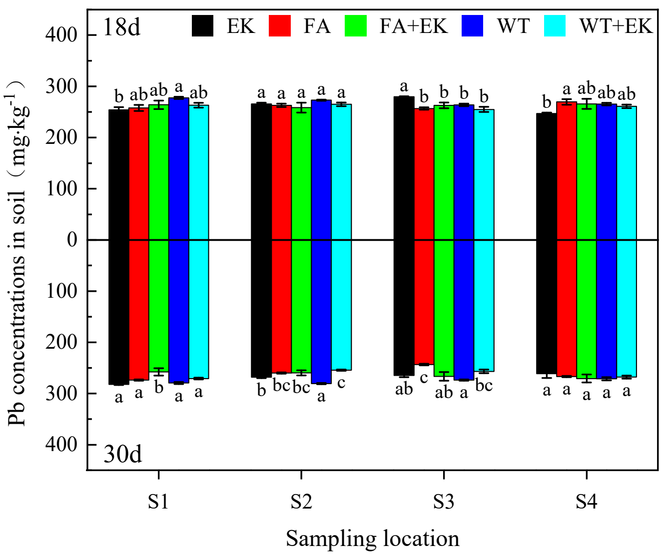

Figure 3. The effects of different treatments on Pb concentration in soil at different sampling points of 18 days and 30 days. FA and WT indicated that tall fescue and wheat were sown separately without DC. FA + EK and WT + EK indicated that tall fescue and wheat were sown under DC conditions. Different lowercase letters on the column indicated significant differences between different treatments ($p < 0.05$).

3.2. Accumulation and Translocation of Pb in Tall Fescue and Wheat

Compared with the accumulation of Pb in tall fescue and wheat, BCF and TF can better indicate the accumulation and transport of Pb in tall fescue and wheat. After uptake of Pb for 18 d, the BCF of tall fescue at S2, S3, and S4 in the FA + EK treatment group increased significantly, by 10.6%, 9.3%, and 19.1%, respectively, compared with that of the FA treatment group (Figure 4a). Meanwhile, the BCF of wheat seedlings at S1 and S2 in the WT + EK treatment group increased significantly, by 41.7% and 51.1%, respectively, compared with that in the WT treatment group. After 30 d, the BCF of tall fescue at S1, S2, and S3 in the FA + EK treatment groups increased significantly, by 236%, 102.7%, and 160%, respectively, compared with that of the FA treatment group (Figure 4b). Furthermore, the BCF of wheat at S2, S3, and S4 in the WT + EK treatment group increased significantly, by 76.7%, 30.3%, and 57.6%, respectively, compared with that in the WT treatment group.

After electrification, the enrichment coefficient of wheat was higher than that of tall fescue at 18 d. The DC exchange electric field significantly increased the Pb enrichment coefficient of tall fescue and wheat seedlings. However, at 30 d, the enrichment coefficient of tall fescue was higher than that of wheat. Considering the long-term removal effect, tall fescue is more suitable than wheat for remediating Pb-contaminated soil.

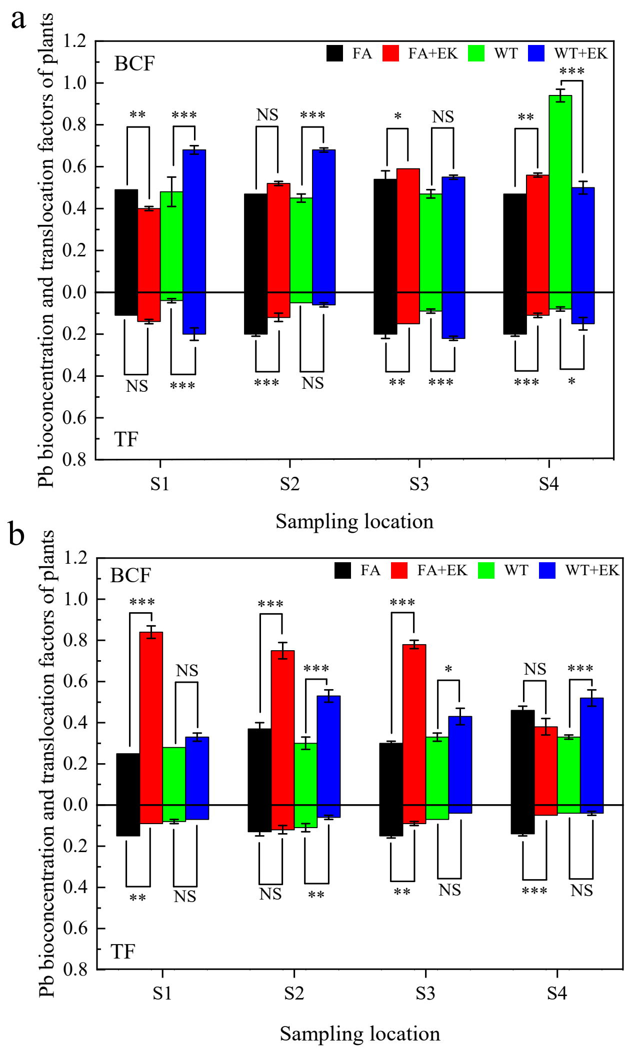

Figure 4. Effects of different treatments on plant bioconcentration factor and translocation factor at different sampling points of 18 days (**a**) and 30 days (**b**). BCF: bioconcentration factor. FT: translocation factor. FA and WT indicated that tall fescue and wheat were sown separately without DC. FA + EK and WT + EK indicated that tall fescue and wheat are sown under DC conditions. NS: not significant, *** $p \leq 0.001$; ** $p \leq 0.01$; * $p \leq 0.05$.

Pb in the soil is extracted by plant roots and transported to the aboveground part; its transport capacity is restricted by many factors, and the accumulation of heavy metals in plants is related to a variety of defense mechanisms [32]. After treatment for 18 d, the TF of tall fescue at S2, S3, and S4 in the FA + EK treatment groups reduced significantly, by 40%, 25%, and 45%, respectively, compared with that of the FA treatment group. The TF of wheat seedlings at S1, S3, and S4 in the WT + EK treatment group increased significantly, by 400%, 144.4%, and 87.5%, respectively, compared with that of the WT treatment group (Figure 4a). After treatment for 30 d, the TF of tall fescue at S1, S2, and S3 in the FA + EK treatment group reduced significantly, by 40%, 40%, and 64.3%, respectively, compared with that of the FA treatment group. The TF of wheat at S2 position in the WT + EK treatment group reduced significantly, by 45.5%, compared with that of the WT treatment group (Figure 4b).

The upward transport of Pb from wheat seedling roots to shoots was greatly promoted by electric stimulation for 18 d. However, Pb transport in both plants was restrained after 18 d of electric stimulation. The upward transport coefficient of Pb from the roots to the shoots in both plants was obviously restrained by the exchange electrode due to the current change.

3.3. Current Changes

The effect of tall fescue and wheat on the current change during the electro-assisted phytoremediation of heavy-metal-contaminated soil is shown in Figure 5. In an electrokinetic remediation system, the current value is closely related to the direction of the daily exchange electrode of the system. After the exchange electrode was energized for 6 h, the current changed sharply from the cycle set on day 1. In the experimental period of 30 d, the soil moisture content, electroosmotic flow, and current in the soil changed regularly because of physical, chemical, and biological effects (such as electrolysis and evaporation). The values gradually decreased during each cycle. After the electrode was exchanged, the current returned to a higher value. The current changes in the EK, FA + EK, and WT + EK groups were similar to the jagged rule. In the polarity reversal electrical field, since the initial electrical field (from the initial anode to the initial cathode) changed the soil ion concentration distribution, the current density and electrolysis reaction rate in the subsequent reverse electric field (from the initial cathode to the initial anode) changed [18]. During the entire repair process, the currents of the FA + EK and WT + EK treatment groups were significantly higher than that of the EK treatment group. At the end of the repair, the current of the FA + EK and WT + EK treatment groups tended to be gentle, with a value of approximately 40 mA, whereas that of the EK treatment group was approximately 25 mA. This may be due to the adhesion of tall fescue and wheat plant roots to soil ions, which slows down the migration of ions to the cathode and anode and maintains the stability of the system current [33]. This phenomenon is consistent with the results of conductivity measurements in the soil. Plant roots may reduce the migration of soil ions, indicating that plants help to maintain soil EC values to a certain extent. The DC electric field with polarity reversal keeps the soil pH stable, which is beneficial to the normal growth process of tall fescue and wheat. Tall fescue and wheat roots may also increase the current value by increasing soil porosity. Tall fescue and wheat enhanced the current, improved the electrodynamic effect of the system, and made the electrokinetic effect continuously stimulate the metabolic activity of the repair plant. Therefore, planting tall fescue and wheat enhanced the remediation of contaminated soil as electrically assisted plants.

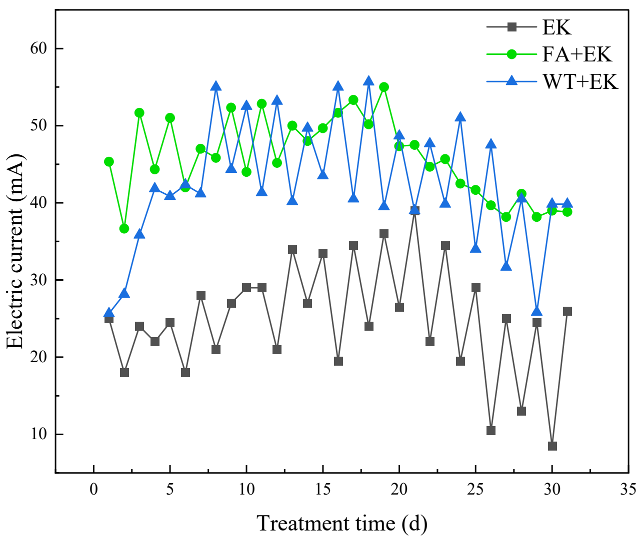

Figure 5. The influence of planting on current change. EK: a treatment unseeded with DC alone. FA + EK and WT + EK indicated that tall fescue and wheat are sown under DC conditions.

3.4. Soil pH and Electrical Conductivity

Soil pH is an important parameter in electrokinetic treatment. The change in the pH was due to the hydrolysis reaction in the electrode area. The pH values of all treatment groups in the Figure were in the range of 7.45–7.71, and the variation range did not exceed one unit amplitude (Figure 6a). It can be seen that the periodic exchange electrode prevented the extreme pH value at both ends of the soil chamber, which was consistent with the research results of Luan et al. [34]. The polarity reversal DC electrical field makes the soil pH change moderate, which was beneficial to plant growth [35]. Slight soil acidification was observed in the treatment group treated with an electric field, mainly because of the faster migration rate of the H^+ ($2H_2O \rightarrow O_2 + 4H^+ + 4e^-$) produced by the anodic reaction in the electrokinetic process compared to that of the OH^- ($2e^- + 2H_2O \rightarrow 2OH^- + H_2$) produced by the cathode [36]. The polarity reversal DC electric field made H^+ ions and OH^- ions generated cyclically on the cathode and anode. After polarity reversal, the H^+ and OH^- produced by the electrolytic reaction could be neutralized. It reduced the soil acidification effect and Pb precipitation, while also avoiding the formation of soil alkaline environment. Since the initial electrolysis reaction changed the distribution of soil ion concentration, the electrolysis reaction rate and current density changed in the subsequent process.

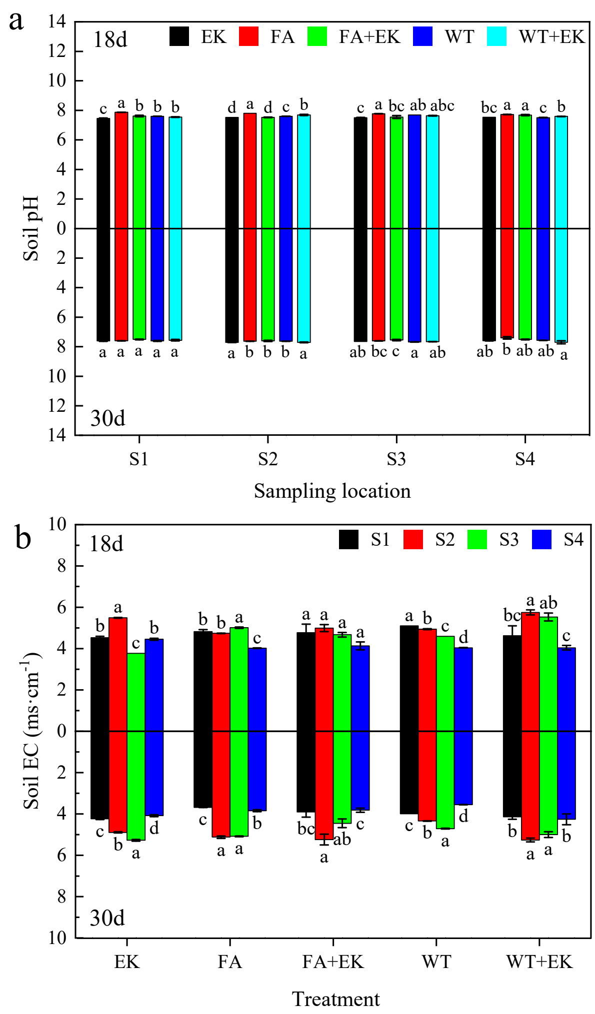

Figure 6. Effects of different treatments on soil pH (**a**) and EC value (**b**) at different sampling points of 18 and 30 days. EK: a treatment unseeded with DC alone. FA and WT indicated that tall fescue and wheat were sown separately without DC. FA + EK and WT + EK indicated that tall fescue and wheat were sown under DC conditions. Different lowercase letters on the column indicated significant differences between different treatments ($p < 0.05$).

Planting tall fescue and wheat in the soil delayed the current decay trend of the electrokinetic remediation system, which was also reflected in the changes in soil EC values (Figure 6b). Soil EC is closely related to charged particles in the soil [21]. After 30 d of electrification, the soil EC values at the S1 and S4 positions in the EK, FA + EK, and WT + EK treatment groups were significantly lower than those at the S2–S3 positions ($p < 0.05$). The negative and positive ions in the soil moved to the positive and negative electrodes, respectively, and under the action of the electrical field during the electrification process, the mobile ions in the soil gradually decreased and the conductivity weakened. The polarity reversal DC electrical field may cause the dissolved ions to migrate to the soil's middle section. After 30 d, the EC values of soil at the S1 and S4 positions in the FA and WT treatment groups were significantly lower than those in the EK treatment group ($p < 0.05$), indicating that the electrolyte in the electrode chamber on both sides of the soil chamber had a major influence on the EC value of the soil. The charged particles in the soil at positions S1 and S4 entered the electrolyte and also had a major influence on the reduction of the EC value by a large amount. The change in soil EC values in S1 and S4 of the WT + EK treatment group was lower than that in the EK treatment group, indicating that plants could help maintain soil EC values to a certain extent [37]. The results showed that plant roots may reduce the migration of soil ions. Plant roots may increase the current value by reducing the migration of soil ions and increasing soil porosity.

4. Conclusions

The DC exchange electric field significantly increased the uptake and accumulation of Pb by tall fescue and wheat seedlings owing to improved soil pH and conductivity. The wheat seedlings showed a better remediation potential in a short period (18 d), whereas tall fescue seemed more suitable for repairing the Pb-contaminated soil over a long period (30 d). Tall fescue was more suitable than wheat seedlings for use as a remediation plant for Pb-contaminated soil. The upward transport coefficient of Pb from the roots to the shoots in both plants was obviously restrained by the exchange electrode due to the current change. Thus, the DC exchange electrode field is conducive to both phytoremediation efficiency and agricultural product safety. The combined remediation technology of exchange electric fields and phytoremediation has strong application prospects for the treatment of Pb-contaminated soil. Future research should examine the selection and testing of promising hyperaccumulators for electrokinetic-assisted phytoremediation of Pb-contaminated soil.

Author Contributions: Conceptualization, H.M. and A.M.; methodology, N.A.; data curation, H.M. and L.J.; writing—original draft preparation, H.M.; investigation, N.L.; software, Y.H.; writing—review and editing, Y.S.; project administration, A.M. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Natural Science Foundation of Xinjiang Uygur Autonomous Region of China [grant No. 2021D01C049].

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The datasets are available from the corresponding author on reasonable request.

Conflicts of Interest: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

1. Li, X.P.; Wang, J.W.; Zhang, Y.N. Review of toxic lead in urban soil and exposure risk to children. *Med. Geogr.* **2016**, *37*, 85–92.
2. Vardhan, K.H.; Kumar, P.S.; Panda, R.C. A review on heavy metal pollution, toxicity and remedial measures: Current trends and future perspectives. *J. Mol. Liq.* **2019**, *290*, 111–197. [CrossRef]
3. Ashraf, S.; Ali, Q.; Zahir, Z.A.; Ashraf, S.; Asghar, H.N. Phytoremediation: Environmentally sustainable way for reclamation of heavy metal polluted soils. *Ecotoxicol. Environ. Saf.* **2019**, *174*, 714–727. [CrossRef]

4. Zhang, Y.H.; Hou, D.Y.; O'Connor, D.; Shen, Z.T.; Shi, P.L.; Ok, Y.S.; Tsang, D.C.W.; Wen, Y.; Luo, M.N. Lead contamination in Chinese surface soils: Source identification, spatial-temporal distribution and associated health risks. *Crit. Rev. Environ. Sci. Technol.* **2019**, *49*, 1386–1423. [CrossRef]
5. Niu, H.; Wang, Z.L.; Song, J.N.; Long, A.G.; Cao, M.; Luo, J. Cadmium subcellular distribution and chemical form in Festuca arundinacea in different intercropping systems during phytoremediation. *Chemosphere* **2021**, *276*, 130–137. [CrossRef]
6. Jin, Z.; Deng, S.; Wen, Y.; Jin, Y.; Pan, L.; Zhang, Y.; Black, T.; Jones, K.C.; Zhang, H.; Zhang, D. Application of Simplicillium chinense for Cd and Pb biosorption and enhancing heavy metal phytoremediation of soils. *Sci. Total Environ.* **2019**, *697*, 134–148. [CrossRef]
7. Hong, Y.; Heerink, N.; Jin, S.Q.Q.; Berentsen, P.; Zhang, L.Z.; van der Werf, W. Intercropping and agroforestry in China—Current state and trends. *Agric. Ecosyst. Environ.* **2017**, *244*, 52–61. [CrossRef]
8. Liu, W.Q.; Zhu, F.; Ma, S.Y. Research Progress on the Electro-kinetic Remediation of Soil Polluted by Heavy Metal. *Saf. Environ. Eng.* **2015**, *22*, 6.
9. Azhar, U.; Ahmad, H.; Shafqat, H.; Babar, M.; Shahzad Munir, H.M.; Sagir, M.; Arif, M.; Hassan, A.; Rachmadona, N.; Rajendran, S.; et al. Remediation techniques for elimination of heavy metal pollutants from soil: A review. *Environ. Res.* **2022**, *214*, 113918. [CrossRef] [PubMed]
10. Begum, Z.A.; Rahman, I.M.M.; Tate, Y.; Sawai, H.; Maki, T.; Hasegawa, H. Remediation of toxic metal contaminated soil by washing with biodegradable aminopolycarboxylate chelants. *Chemosphere* **2012**, *87*, 1161–1170. [CrossRef]
11. Song, P.; Xu, D.; Yue, J.; Ma, Y.; Dong, S.; Feng, J. Recent advances in soil remediation technology for heavy metal contaminated sites: A critical review. *Sci. Total Environ.* **2022**, *838*, 156417. [CrossRef] [PubMed]
12. Liu, L.; Li, W.; Song, W.; Guo, M. Remediation techniques for heavy metal-contaminated soils: Principles and applicability. *Sci. Total Environ.* **2018**, *633*, 206–219. [CrossRef] [PubMed]
13. Awa, S.H.; Hadibarata, T. Removal of Heavy Metals in Contaminated Soil by Phytoremediation Mechanism: A Review. *Water Air Soil Pollut.* **2020**, *231*, 47. [CrossRef]
14. Ali, H.; Khan, E.; Sajad, M.A. Phytoremediation of heavy metals-Concepts and applications. *Chemosphere* **2013**, *91*, 869–881. [CrossRef] [PubMed]
15. Elbehiry, F.; Elbasiouny, H.; Ali, R.; Brevik, E.C. Enhanced Immobilization and Phytoremediation of Heavy Metals in Landfill Contaminated Soils. *Water Air Soil Pollut.* **2020**, *231*, 204. [CrossRef]
16. Liu, Y.; Niu, T.; Li, T.; Jiang, M.; Xiong, J.; Li, B.; Zhan, F.; He, Y. Characteristics and opportunities of electrokinetic-assisted phytoremediation of heavy metal contaminated soil. *Chem. Ind. Eng. Prog.* **2020**, *39*, 5252–5265.
17. Yeung, A.T.; Gu, Y.Y. A review on techniques to enhance electrochemical remediation of contaminated soils. *J. Hazard. Mater.* **2011**, *195*, 11–29. [CrossRef]
18. Yuan, L.; Guo, P.; Guo, S.; Wang, J.; Huang, Y. Influence of electrical fields enhanced phytoremediation of multi-metal contaminated soil on soil parameters and plants uptake in different soil sections. *Environ. Res.* **2021**, *198*, 111290. [CrossRef]
19. Aboughalma, H.; Bi, R.; Schlaak, M. Electrokinetic enhancement on phytoremediation in Zn, Pb, Cu and Cd contaminated soil using potato plants. *J. Environ. Sci. Health Part A-Toxic/Hazard. Subst. Environ. Eng.* **2008**, *43*, 926–933. [CrossRef]
20. Lim, J.M.; Salido, A.L.; Butcher, D.J. Phytoremediation of lead using Indian mustard (*Brassica juncea*) with EDTA and electrodics. *Microchem. J.* **2004**, *76*, 3–9. [CrossRef]
21. Cang, L.; Wang, Q.Y.; Zhou, D.M.; Xu, H. Effects of electrokinetic-assisted phytoremediation of a multiple-metal contaminated soil on soil metal bioavailability and uptake by Indian mustard. *Sep. Purif. Technol.* **2011**, *79*, 246–253. [CrossRef]
22. Siyar, R.; Doulati Ardejani, F.; Farahbakhsh, M.; Norouzi, P.; Yavarzadeh, M.; Maghsoudy, S. Potential of Vetiver grass for the phytoremediation of a real multi-contaminated soil, assisted by electrokinetic. *Chemosphere* **2020**, *246*, 125802. [CrossRef] [PubMed]
23. Li, J.X.; Zhang, J.; Larson, S.L.; Ballard, J.H.; Guo, K.; Arslan, Z.; Ma, Y.H.; Waggoner, C.A.; White, J.R.; Han, F.X.X. Electrokinetic-enhanced phytoremediation of uranium-contaminated soil using sunflower and Indian mustard. *Int. J. Phytoremediat.* **2019**, *21*, 1197–1204. [CrossRef] [PubMed]
24. Li, J.; Wen, Y.; Li, X.; Li, Y.; Yang, X.; Lin, Z.; Song, Z.; Cooper, J.M.; Zhao, B. Soil labile organic carbon fractions and soil organic carbon stocks as affected by long-term organic and mineral fertilization regimes in the North China Plain. *Soil Tillage Res.* **2018**, *175*, 281–290. [CrossRef]
25. Li, T.; Mu, T.; Liu, G.; Yang, X.; Zhu, G.; Shang, C. A Method of Soil Moisture Content Estimation at Various Soil Organic Matter Conditions Based on Soil Reflectance. *Remote Sens.* **2022**, *14*, 2411. [CrossRef]
26. Bache, B.W. Barium isotope method for measuring cation-exchange capacity of soils and clays. Journal of the Science of Food and Agriculture, 22, 169–171. *J. Sci. Food Agric.* **1970**, *21*, 169–171. [CrossRef]
27. Chiriac, E.R.; Chiţescu, C.L.; Sandru, C.; Geană, E.-I.; Lupoae, M.; Dobre, M.; Borda, D.; Gird, C.E.; Boscencu, R. Comparative Study of the Bioactive Properties and Elemental Composition of Red Clover (*Trifolium pratense*) and Alfalfa (*Medicago sativa*) Sprouts during Germination. *Appl. Sci.* **2020**, *10*, 7249. [CrossRef]
28. Azeez, J.O.; Olowoboko, T.B.; Bada, B.S.; Odedina, J.N.; Onasanya, O.O. Evaluation of soil metal sorption characteristics and heavy metal extractive ability of indigenous plant species in Abeokuta, Nigeria. *Int. J. Phytoremediat.* **2020**, *22*, 872–884. [CrossRef] [PubMed]

29. Rutkowska, B.; Szulc, W.; Blaszczak, E.; Kazberuk, W.; Ptasinski, D. Restoration of marginal soils polluted with heavy metals to agricultural production. *J. Soil Water Conserv.* **2020**, *75*, 610–616. [CrossRef]
30. Zhang, W.H.; Zhang, L.L.; Mao, S.Y.; Qiu, R.L. Migration and Stabilization of Multiple Heavy Metals in an Aged Contaminated Soil under a Constant Voltage Electric Field. *Soil Sediment Contam.* **2014**, *23*, 540–556. [CrossRef]
31. Putra, R.S.; Ohkawa, Y.; Tanaka, S. Application of EAPR system on the removal of lead from sandy soil and uptake by Kentucky bluegrass (*Poa pratensis* L.). *Sep. Purif. Technol.* **2013**, *102*, 34–42. [CrossRef]
32. Mertens, J.; Vervaeke, P.; Schrijver, A.D.; Luyssaert, S. Metal uptake by young trees from dredged brackish sediment: Limitations and possibilities for phytoextraction and phytostabilisation. *Sci. Total Environ.* **2004**, *326*, 209–215. [CrossRef]
33. Chirakkara, R.A.; Reddy, K.R.; Cameselle, C. Electrokinetic Amendment in Phytoremediation of Mixed Contaminated Soil. *Electrochim. Acta* **2015**, *181*, 179–191. [CrossRef]
34. Luan, Y.; Xu, J.; Zhou, J.; Wang, H.; Han, F.; Wang, K.; Lv, Y. Migration and Removal of Labile Cadmium Contaminants in Paddy Soils by Electrokinetic Remediation without Changing Soil pH. *Int. J. Environ. Res. Public. Health* **2022**, *19*, 3812. [CrossRef]
35. Bi, R.; Schlaak, M.; Siefert, E.; Lord, R.; Connolly, H. Influence of electrical fields (AC and DC) on phytoremediation of metal polluted soils with rapeseed (*Brassica napus*) and tobacco (*Nicotiana tabacum*). *Chemosphere* **2011**, *83*, 318–326. [CrossRef] [PubMed]
36. O'Connor, C.S.; Lepp, N.W.; Edwards, R.; Sunderland, G. The combined use of electrokinetic remediation and phytoremediation to decontaminate metal-polluted soils: A laboratory-scale feasibility study. *Environ. Monit. Assess.* **2003**, *84*, 141–158. [CrossRef]
37. Kravkaz Kuscu, I.S. Certain soil characteristics and light conditions of enzyme activities and variance conditional to plant type. *Environ. Monit. Assess.* **2020**, *192*, 229. [CrossRef]