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Synergistic Electrokinetic Biochar Remediation for Heavy Metal Immobilization and Agroecological Treatment of Contaminated Soils

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Abstract

Contamination of agricultural and industrial lands by environmentally dangerous metals (heavy metals) such as Cd, Pb, Cu, and Zn poses a major threat to food security, ecosystem balance, and the agrobiological capability of soils in the arid regions of Kazakhstan. This research thoroughly assessed the efficiency of the combined application of electrokinetic (EK) remediation and pyrolyzed organic matter (biochar) across three distinct soil types - Light gray soil, Light Chestnut Soil, and chernozem (black) soil. The fundamental soil characteristics (pH, trace elements, organic matter, macronutrients, cation exchange capacity, exchangeable cations, particle size distribution) were identified per international guidelines.

The remediation process was divided into two phases: (1) EK treatment (30 V, 12 hours, 0.01 M citric acid), and (2) biochar augmentation (0, 15, 25, 35 t/ha). Findings indicated that the EK treatment alone diminished the mobile fractions of heavy metals by approximately 20–60%, whereas the combination of EK + biochar enhanced this effectiveness to 60–95%. The most significant immobilization was recorded for Cu and Zn: in Light gray soil and chernozem soils, the decline in the mobile form of Zn reached 76–80%, and for Cu — 61–95%. Pb and Cd also demonstrated substantial reductions, being immobilized within the range of 65–80%.

Differences between soil types were revealed: in chernozem soils, biochar bound heavy metals most intensely, leading to a sharp decrease in mobile forms; whereas in Light gray soil and Light Chestnut soils, the immobilization efficiency increased proportionally with the dose applied.

A multifactorial ANOVA revealed a substantial impact of biochar dosage, soil type, and pollution severity on all assessed parameters ($p < 0.001$; η^2 ranged from 0.75 to 0.93). Tukey's Honestly Significant Difference (HSD) test further confirmed that all tested biochar doses (0, 15, 25, and 35 t/ha) produced statistically significant variations. Phytomeliorant yields showed a considerable enhancement across all soil types, with plant numbers rising 3 to 10-fold, plant heights increasing 2 to 5-fold, and productivity index (PI) peaking at approximately 7000 to 7500 in chernozem soils. This indicates that biochar reduces the phytotoxicity of heavy metals.

Overall, EK + biochar technology is a highly effective, synergistic approach for restoring soils contaminated with heavy metals to an environmentally safe condition and for recovering their agrobiological productivity under various natural-regional conditions of Kazakhstan.

Keywords: electrokinetic remediation, biochar, heavy metals, soil fertility, immobilization, phytoremediation, soils of Kazakhstan.

1. Introduction

Heavy metal pollution of various soil types, which is of great importance in agriculture, is currently one of the most serious global problems affecting food security, ecosystem stability, and human health. According to international estimates, 14-17% of the soils used for agriculture in the world are contaminated with at least one heavy metal (for example, Cd, Pb, Zn, and Cu) at levels exceeding the threshold limit [1]. The stability of heavy metals, their resistance to biodegradation, and long-term storage in the soil profile place them among sustainable environmental toxicants [2]. Numerous studies have shown that Cd and Pb, in particular, have high toxicity properties for the environment and can undergo bioaccumulation in soil, plant, and human systems [3].

In the conditions of Kazakhstan, the accumulation of environmentally hazardous metals is strongly influenced by natural and climatic conditions, geochemical structure, industrial pressure, agricultural loads, and irrigation systems. Periodic irrigated agriculture in the southern and southwestern regions, especially in Kyzylorda and Turkestan, and degradation processes in the Aral Sea basin, especially salinization, have reduced soil resistance to negative environmental factors, increased soil pollution, and reduced land productivity. In addition, the accumulation of toxic metals in soils poses significant ecological and public health risks by negatively affecting microbial activity, agricultural productivity, ecosystem functions, and human health [4]. According to studies conducted in the Syrdarya Valley, Cd, Pb, Cu, and Zn determine the patterns of vertical migration, their accumulation coefficients, and high levels of phytotoxicity [5]. Even in urbanized areas, environmentally hazardous metals significantly reduce the sanitary and hygienic qualities of soils and threaten public health [6,7].

Since more than 70% of Kazakhstan's industrial facilities are located in desert and semi-desert zones, soil pollution with toxic metals harmful to the environment has become an important and urgent environmental problem in the country [8]. Therefore, the fact that the restoration of these lands is very long-term and requires modern, rational remediation methods has become relevant today.

The movement and change in soil quality under the influence of environmentally harmful metals on Earth is influenced by typological and morphogenetic features. For example, chernozem (black soil) soils, rich in humus with high fertility and dominated by calcium in the soil exchange complex, can effectively bind ionic and complex forms of metals, thereby reducing their bioavailability [9]. On the contrary, weakly structured soils with low organic content and light mechanical composition-for example, light gray soil and light chestnut soil-allow high amounts of mobile metal fractions to accumulate and, at the same time, increase the risk of plants absorbing metals into themselves [10]. Therefore, soil contamination with various toxic metals and their recovery depend on the type of soil and fertility parameters.

Several technologies have been used to restore soils contaminated with environmentally hazardous metals, including traditional recovery methods. Among them, electrokinetic (EK) recovery has become one of the methodologies of great interest in soil remediation in recent years, and EK technology has been recognized as highly effective for soils with clay mechanical composition and low conductivity due to its ability to move ions under an electric field [11]. In recent years, sustainable remediation frameworks have increasingly emphasized environmentally friendly and integrated approaches capable of simultaneously reducing metal mobility and restoring soil ecological functionality[12]. Many studies on this methodology are revealed in the results of the study that the EK recovery methodology has a high efficiency in various types of soils [13]. However, the independent application of EK remediation still faces several limitations, including pH polarization near the cathode, precipitation of metal hydroxides, secondary contamination risks, and insufficient recovery of soil biological functions. [14].

In this regard, hybrid technologies have been widely developed that combine EK with additional materials such as chelating agents, reactive barriers, nanomaterials, and biochar to increase efficiency [15]. Biomass subjected to carbon-rich pyrolysis due to the thermal decomposition of plant byproducts shows a fairly accessible surface area (ASA), which facilitates the effective adsorption and immobilization of harmful environmental metals along with micro- and mesoporous structural characteristics and many functional groups [16].

The use of biochar in the post-treatment period by the remediation method of contaminated soils regulates the pH of the soil, increases the CEC, improves the soil structure, increases resistance to pollutants that negatively affect the soil, and thus increases the biological activity of the soil and plant growth [17]. Amendment-based remediation approaches using biochar have shown promising results for Pb and Cd immobilization while simultaneously improving soil fertility and phytoremediation performance[18–20]. Remediation of soils based on total pyrolysis biomass stabilizes its long-term sorption capacity and forms complex structural bonds with metals [21].

In the remediation of highly polluted soils, the combination of EK and biochar has a synergistic effect: EK mobilizes environmentally hazardous metals, and pyrolysis biomass permanently immobilizes them. Such technologies have shown high efficiency on multicomponent contaminated soils, especially for soils in harsh, arid regions with low organic matter climates [22].

At the same time, the assessment of the agrobiological quality of soils after recovery through such combined methods, the study of plant productivity, and biomass formation remain insufficiently solved in modern studies. It has been confirmed that pyrolysis biomass increases plant resistance to stress factors and reduces the phytotoxic effect of heavy metals by improving the physical properties of the soil and increasing the biological activity of the soil [23]. Therefore, in soils with the introduction of organic and carbon biomeliorants, the growth dynamics of phytoremediation crops and the development of the root system are significantly improved, allowing the soil to fully form [24]. Therefore, the efficiency and importance of the restoration of soils of Kazakhstan with a dry climate, highly polluted with low-fertility heavy metals, by sequential use of 3-stage methods using EK, biochar, and phytomeliorants is high.

Nevertheless, most previous studies have primarily focused on metal removal efficiency or short-term physicochemical changes, while the agrobiological recovery of remediated soils, including plant productivity, phytomeliorative performance, and restoration of soil ecological functions, remains insufficiently investigated.

Despite the growing number of studies on electrokinetic remediation and biochar-assisted immobilization of heavy metals, limited information is available regarding the combined application of these technologies across soils with contrasting physicochemical properties. In particular, the mechanisms governing the interaction between electrokinetic mobilization and biochar-induced stabilization under different soil conditions remain insufficiently understood. Furthermore, the influence of biochar dosage on remediation efficiency and ecological recovery has not been comprehensively evaluated for Kazakhstan soils.

Therefore, the objective of this study was to evaluate the effectiveness of combined electrokinetic remediation and biochar application for reducing the mobility and bioavailability of Cd, Pb, Cu, and Zn in contaminated soils differing in texture, organic matter content, and agrochemical properties.

It was hypothesized that:

(1) the combined application of electrokinetic treatment and biochar would produce a synergistic effect, resulting in greater reductions in heavy-metal mobility compared with individual treatments;

(2) the degree of heavy-metal removal and immobilization would depend on soil type and biochar application dose due to differences in soil physicochemical characteristics and sorption capacity; and

(3) decreasing heavy-metal bioavailability would significantly improve phytoremediation plant growth and contribute to restoration of soil ecological functions.

2. Materials and methods

2.1. Collection of soil samples

In this study, three different soil types were used to evaluate the effectiveness of electrokinetic remediation combined with biochar and phytomeliorant applications for the restoration of heavy metal-contaminated soils. Prior to the experiments, the collected soil samples were pretreated under laboratory conditions following standard soil preparation protocols commonly reported in electrokinetic remediation studies.

Initially, visible plant residues, stones, and other coarse impurities were manually removed from the soils. The samples were then air-dried at room temperature to preserve the natural physicochemical properties of the soil and to minimize changes in heavy metal speciation during preparation [25]. After drying, the soils were homogenized and sieved through a 2 mm mesh sieve to obtain uniform particle size distribution. Sieving is a widely accepted pretreatment step in soil remediation studies because it improves sample homogeneity and ensures more uniform distribution of contaminants within the soil matrix [26]. Furthermore, homogeneous fine particles enhance ion migration and electroosmotic flow during electrokinetic treatment processes [27].

Artificial contamination of the soils was performed using aqueous solutions of heavy metal salts, including Cd, Pb, Cu, and Zn compounds. The required concentrations of metal salts were prepared using deionized water and gradually introduced into the soil by spraying and mechanical mixing to ensure uniform adsorption of heavy metals throughout the soil matrix. Similar artificial spiking procedures have been extensively applied in electrokinetic remediation experiments to simulate contaminated soils under controlled laboratory conditions [27].

Following contamination, the soils were thoroughly mixed using a mechanical stirrer and incubated in sealed polyethylene containers for 7–14 days to establish equilibrium between heavy metal ions and soil particles. This aging process allows the metals to interact with soil colloids and organic matter, thereby mimicking naturally contaminated soils more realistically [25]. During incubation, soil moisture content was maintained at approximately 60–70% of water holding capacity to facilitate metal mobility and uniform contaminant distribution [26].

After the incubation period, biochar and phytomeliorant amendments were incorporated into the contaminated soils prior to electrokinetic treatment. Biochar application has been reported to reduce heavy metal mobility through adsorption and surface complexation mechanisms, while phytoremediation-assisted electrokinetic systems can enhance metal uptake and removal efficiency from contaminated soils [28, 29].

2.2. Determination of initial agrochemical and chemical properties of soil

Initial agrochemical and physicochemical properties of the soil samples were determined using standard methods commonly applied in soil science, agrochemistry, and environmental studies, in accordance with Kazakhstan regulatory documents and relevant GOST standards.

Soil pH was measured potentiometrically in soil suspensions prepared at a soil-to-solution ratio of 1:2.5 (w/v) using distilled water and 1 M KCl solution, following standard agrochemical procedures [30]. Measurements were performed using a calibrated glass-electrode pH meter after equilibration of the suspensions.

The granulometric composition (particle-size distribution) of soils was determined using the Kachinsky pipette method in accordance with GOST 12536-2014 [31]. The analysis included the separation and quantification of major soil particle-size fractions.

Soil organic matter content was determined according to GOST 26213-91 [32]. Humus content and its fractional composition were assessed using the Ponomareva and Plotnikova method. The elemental composition of humic acids was analyzed using an automated elemental analyzer.

Easily hydrolyzable nitrogen was determined using the Tyurin–Kononova method [33]. All laboratory analyses were performed in triplicate to ensure analytical accuracy and reproducibility.

Total carbonate ($CaCO_3$) content (%) was determined using the gas-volumetric calcimetric method based on measuring the volume of CO_2 released during reaction with hydrochloric acid. Total nitrogen was measured using the Kjeldahl method after sulfuric acid mineralization with a catalyst [34]. Ammonia released during steam distillation was absorbed in boric acid and quantified by titration. Total potassium content was determined according to the Michigan method following classical agrochemical procedures [35]. Total Phosphorus (P_2O_5) and Potassium (K_2O) - Soils underwent acid mineralization following GOST 26207 and modified Mehlich procedures [36]. Phosphorus was measured colorimetrically, potassium-by flame photometry or atomic absorption spectroscopy (AAS). Mobile Phosphorus (P_2O_5) and Potassium (K_2O) - Was determined using the Chirikov method and potassium-by ammonium acetate extraction (CH_3COONH_4) [37]. Exchangeable cations and cation exchange capacity (CEC) of the soil samples were determined using the ammonium acetate extraction method in accordance with ISO 23470 [38]. Soil samples were extracted with ammonium acetate solution to replace exchangeable cations adsorbed on the soil surface.

Exchangeable calcium (Ca^{2+}) and magnesium (Mg^{2+}) contents were determined by ethylenediaminetetraacetic acid (EDTA) titration according to GOST 26487-85 [39]. Exchangeable sodium (Na^{+}) and potassium (K^{+}) concentrations were measured using flame atomic absorption spectrometry (FAAS).

2.3. Electrokinetic (EK) remediation method

Electrokinetic treatment to reduce the mobile fraction of heavy metals was performed following the work done by Reddy & Cameselle [40].

The electrokinetic remediation experiments were conducted using a laboratory-scale setup consisting of a soil chamber connected to a direct current (DC) power supply. A schematic diagram of the system is presented in Fig. 1.

Inert graphite electrodes were used as both the anode and cathode to minimize secondary contamination and electrode corrosion. The electrodes were positioned at opposite ends of the soil chamber, with the anode connected to the positive terminal and the cathode to the negative terminal of the power supply.

A constant voltage of 30 V was applied across the electrodes, generating an electric field that facilitated electromigration of charged species and electroosmotic flow through the soil matrix. The direction of ion transport occurred from the anode toward the cathode, consistent with electrokinetic transport mechanisms.

To maintain stable electrochemical conditions, the electrode compartments were periodically monitored, and the electrolyte solution (0.01 M citric acid) was used to enhance metal desorption and mobility within the soil.

The applied voltage of 30 V was chosen as an optimal value reported in previous electrokinetic remediation studies, providing sufficient electric potential gradient for ion migration while avoiding excessive energy consumption, overheating, and soil drying [40]. Lower voltages (<20 V) were found to be less effective in mobilizing heavy metals, while higher voltages (>40 V) may lead to undesirable side effects such as pH extremes and electrode degradation.

The treatment duration of 12 hours was selected based on preliminary trials [11], where shorter durations resulted in insufficient metal removal, while longer durations did not significantly increase efficiency but increased energy costs.

Citric acid acts as an environmentally safe chelating agent, enhancing the mobility and subsequent removal of metals [41]. After EK treatment, the concentrations of mobile, environmentally hazardous metals (Cd, Cu, Pb, and Zn) were reduced.

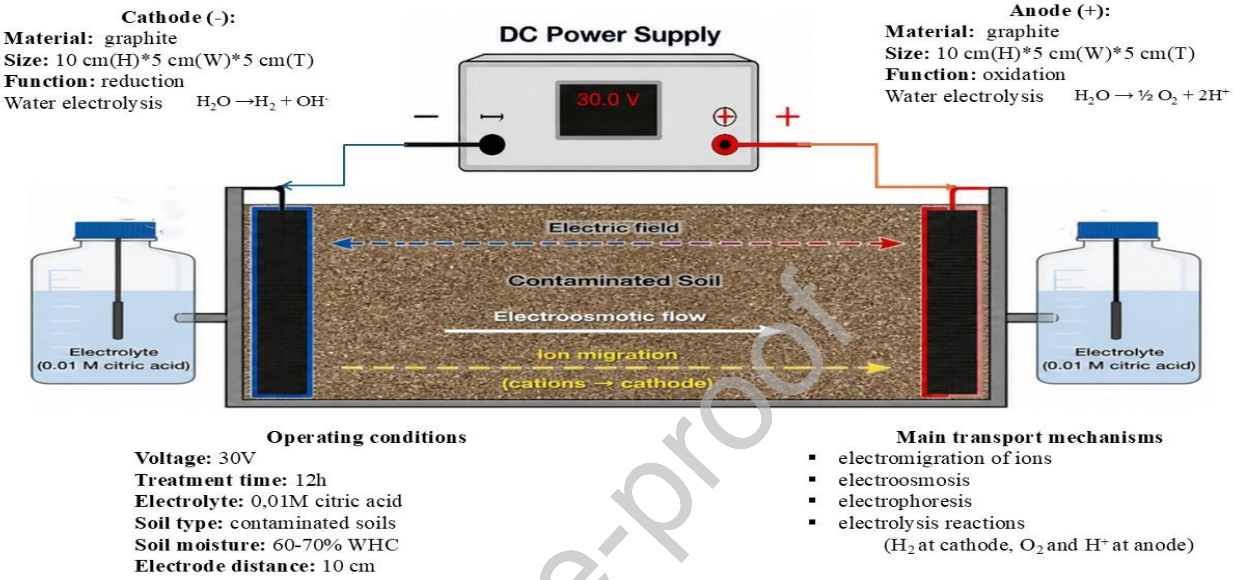

Figure 1. Schematic diagram of the electrokinetic remediation setup

2.4. Methodological characteristics of the biomass pyrolysis process

Pyrolyzed biomass (biochar) was produced by slow pyrolysis of rice husks at 450 °C for 8 h under limited oxygen conditions, following the standards of Lehmann & Joseph (2015). The resulting biochar exhibited high porosity and abundant functional groups (–COOH, –OH), enhancing its heavy-metal adsorption capacity [42]. An increase in biochar application rates (0, 15, 25, and 35 t/ha) resulted in a consistent reduction in the mobile fractions of heavy metals (Cd, Pb, Cu, and Zn) and a significant improvement in plant productivity across all studied soil types. Overall, the immobilization efficiency of heavy metals increased proportionally with biochar dosage, indicating a clear dose-dependent trend within the tested range. This is primarily attributed to the enhanced sorption capacity and surface functional groups of biochar, which promote complexation, precipitation, and adsorption of metal ions in the soil matrix [43].

Although the general trend is dose-dependent and near-linear in the studied range, slight deviations from strict linearity were observed due to differences in soil physicochemical properties such as pH, organic matter content, cation exchange capacity, and texture. In Chernozem soils, higher native organic matter and buffering capacity led to stronger baseline metal immobilization and a faster approach to saturation of biochar sorption sites. In contrast, Light Gray and Light Chestnut soils exhibited a more pronounced response to increasing biochar doses due to their lower natural sorptive capacity [44, 45].

When combined with electrokinetic (EK) remediation, biochar demonstrated a synergistic effect, significantly enhancing the immobilization of heavy metals, with reductions in mobile fractions reaching 60–95%. The EK process facilitates the mobilization of metal ions, while biochar subsequently stabilizes them through adsorption and precipitation mechanisms, resulting in a highly efficient integrated remediation system [11, 46].

The reduction in heavy metal bioavailability led to a marked decrease in phytotoxicity, which in turn significantly improved plant growth parameters. Across all soil types, plant density increased 3–10 times, plant height increased 2–5 times, and the productivity index (PI) reached its maximum values (approximately 7000–7500) in Chernozem soils. These results confirm that biochar application enhances soil fertility and crop performance by reducing metal stress and improving nutrient availability [47, 48].

Overall, the findings demonstrate that increasing biochar dosage effectively enhances heavy metal immobilization and soil ecological safety, while the combined EK + biochar approach represents a highly efficient remediation strategy for restoring contaminated soils under diverse agroecological conditions in Kazakhstan.

2.5. Calculation of biochar mass added to the reactor

The amount of biochar applied to each experimental reactor was calculated based on the equivalent field application rate using the following equation adapted from Reddy and Cameselle [40]:

$$M = \frac{D \times \rho \times V}{10^6} \qquad \text{(Eq. 1)}$$

where:

M= mass of biochar added to the reactor (g);

D= biochar application dose (t ha^{-1});

ρ= soil bulk density (g cm^{-3});

V= working soil volume in the reactor (cm^3).

This equation was used to convert field-scale biochar application rates into equivalent laboratory-scale quantities suitable for reactor experiments. The conversion factor 10^6accounts for the transformation of tonnes per hectare into grams corresponding to the soil volume used in the reactor system.

The reactor dimensions and soil loading volume were kept constant throughout the experiments to ensure comparability among treatments. Biochar was applied at different dosage levels and thoroughly mixed with the contaminated soil to obtain a homogeneous distribution before the electrokinetic remediation process.

The calculation approach was selected to simulate realistic agricultural or remediation application rates under controlled laboratory conditions. Similar scaling methods for laboratory electrokinetic remediation studies have been reported by Reddy and Cameselle [40].

2.6. Phytomelioration experiment

Three different soil types were sieved through a 2 mm mesh. The fraction passing the sieve was weighed to the required amount, placed into experimental reactors, and subsequently subjected to electrokinetic treatment for heavy-metal removal. After electrokinetic treatment, biochar was incorporated into the soils at different application doses and using different amendment methods. Following biochar incorporation, *Bromus inermis*, *Elymus cristatum*, *Onobrychis viciifolia*, and *Trifolium repens* were sown **(Figure 2).**

The experiment was conducted using a completely randomized factorial design including soil type, remediation stage, and biochar dose as experimental factors. Each treatment was performed in triplicate to ensure reproducibility and analytical reliability.

Figure 2. Phytomelioration experiment.

Growth conditions were maintained under controlled laboratory conditions at 22–24 °C with a 12 h light/12 h dark photoperiod, while soil moisture was maintained at 60% of field capacity throughout the experiment. All reactors were maintained under identical environmental conditions to minimize variability among treatments. These experimental conditions were established according to the FAO Phytoremediation Guide.

2.7. Pure live seed (PLS)

The phytoremediation seed mix was applied as a composite blend containing 30% *Bromus inermis*, 25% *Elymus cristatum*, 35% *Onobrychis viciifolia*, and 10% *Trifolium repens*, with a total seeding rate of 63.11 kg/ha.

PLS was calculated using ISTA (2018) guidelines:

$$\text{PLS (\%)} = (\text{Germination (\%)} \times \text{Purity (\%)}) / 100 \qquad \text{(Eq. 2)}$$

The sowing rate adjusted for PLS:

$$\text{PLS rate (kg/ha)} = \text{Seed rate (kg/ha)} / \text{PLS factor}$$

$$\text{PLS factor} = \text{PLS (\%)} / 100 \qquad \text{(Eq. 3)}$$

- Seed rate (kg/ha): Recommended seeding rate based on species requirements.
- PLS (%): Product of seed purity (%) × germination (%), expressed as percent.
- PLS factor: Decimal fraction representing the viable portion of the seed lot.

2.8. Determination of heavy metals in contaminated soil

The total concentrations of Cd, Pb, Cu, and Zn in contaminated soil samples were determined using the microwave-assisted acid digestion method according to USEPA Method 3051A [49]. Briefly, soil samples were digested using a mixture of concentrated acids in a closed-vessel microwave digestion system to ensure complete extraction of metals from the soil matrix. After digestion, the extracts were filtered and diluted with deionized water to the required volume.

The concentrations of heavy metals in the digested solutions were quantified using inductively coupled plasma mass spectrometry (ICP–MS) or atomic absorption spectroscopy (AAS), depending on metal concentration and instrument availability.

The bioavailable (plant-available) fractions of Cd, Pb, Cu, and Zn were extracted using the diethylenetriaminepentaacetic acid (DTPA) extraction method proposed by Lindsay and Norvell [50]. The extracting solution consisted of:

- 0.005 M DTPA,
- 0.1 M triethanolamine (TEA),
- 0.01 M $CaCl_2$,

with the solution pH adjusted to 7.3.

The soil samples were shaken with the DTPA extracting solution under controlled laboratory conditions, after which the suspensions were filtered. The concentrations of extracted metals were subsequently measured using ICP–MS or AAS. The DTPA extraction method is widely used for assessing the mobile and plant-available fractions of Cd^{2+}, Pb^{2+}, Cu^{2+}, and Zn^{2+} in contaminated soils because it effectively simulates metal availability under natural soil conditions.

2.9. Statistical analysis

All experiments and analytical measurements were performed in triplicate, and the results are presented as mean ± standard deviation (SD). Prior to statistical analysis, the data were checked for normality and homogeneity of variance.

Statistical analysis involved descriptive statistics, as well as one-way and factorial analysis of variance (ANOVA), to evaluate the effects of soil type, remediation stage, biochar dose, and their interactions. Tukey’s HSD post-hoc test was used to determine significant differences among treatment means. Correlation analysis was also performed to examine relationships between soil properties, heavy-metal mobility, and plant growth parameters.

Experimental conditions were maintained constant throughout the study to minimize variability among treatments and ensure analytical reliability. A p-value less than 0.05 was considered statistically significant.

3. Results and discussion

3.1. Baseline physicochemical characteristics of soils (0–20 cm depth)

This study investigated three representative soil types of Kazakhstan-chernozem, light chestnut soil, and light gray soil-differing in organic matter content, nutrient status, exchangeable cations, and particle-size distribution. These parameters are fundamental in determining soil buffering capacity, contaminant mobility, and remediation efficiency, particularly under electrokinetic (EK) treatment systems, where soil texture and mineralogy strongly influence metal transport processes [11].

3.1.1. Chernozem

Chernozem exhibited the most favorable physicochemical properties among the studied soils. The humus content reached 4.11%, indicating a well-developed organic horizon. Total nitrogen (0.170%), phosphorus (0.15%), and potassium (1.4%) reflected a sufficient reserve of macronutrients, while mobile forms (N = 10.93 mg/kg, P_2O_5 = 87.84 mg/kg, K_2O = 225 mg/kg) confirmed moderate nutrient availability (**Figure 3**).

The soil reaction was slightly alkaline (pH 8.4), which is typical for steppe chernozems. Exchangeable bases were dominated by calcium (13.60 mEq/100 g) and magnesium (3.20 mEq/100 g), which play a key role in maintaining aggregate stability and high cation-exchange capacity (CEC) (**Figure 3**). Similar dominance of Ca^{2+} in chernozems has been reported to enhance soil fertility and buffering capacity [51].

The particle-size distribution indicated a heavy loam texture, with silt (0.05–0.01 mm) at 41.06% and clay fraction (<0.001 mm) at 45.96% (**Figure 4**). Such fine-textured soils are characterized by high water-holding capacity and strong adsorption potential for heavy metals, which may reduce contaminant mobility but increase long-term retention [52].

3.1.2. Light chestnut soil

Light chestnut soil demonstrated a moderate fertility level with humus content of 2.40%. Total nitrogen, phosphorus, and potassium were 0.136%, 0.14%, and 0.80%, respectively. Mobile nutrient contents (N = 5.73 mg/kg, P_2O_5 = 76.80 mg/kg, K_2O = 180 mg/kg) were lower than in chernozem, indicating reduced nutrient buffering capacity.

The soil reaction remained alkaline (pH 8.5). Exchangeable cations were also dominated by calcium (13.20 mEq/100 g) and magnesium (2.80 mEq/100 g), while sodium (0.21 mEq/100 g) and potassium (0.13 mEq/100 g) were present in minor amounts (**Figure 3**). The dominance of Ca^{2+} in the exchange complex is typical for chestnut soils and influences aggregation and sorption processes of metal ions [53].

Particle-size analysis revealed 44.18% silt and 41.92% clay fractions, classifying the soil as heavy loam (**Figure 4**). This texture provides high moisture retention and significant adsorption surfaces for contaminants, although slightly lower than in chernozem due to reduced organic matter content [54].

3.1.3. Light gray soil

Light gray soil showed the poorest fertility status among the studied soils. Humus content was only 0.65%, indicating weak organic horizon development. Total nitrogen (0.056%), phosphorus (0.16%), and potassium (0.23%) were significantly lower than in the other soils. However, mobile nutrient forms (N = 36.4 mg/kg, P_2O_5 = 36 mg/kg, K_2O = 236 mg/kg) indicated relatively higher soluble potassium mobility.

The soil reaction was slightly alkaline (pH 8.08). Exchangeable cations included Ca (8.42 mEq/100 g), Mg (4.95 mEq/100 g), Na (0.14 mEq/100 g), and K (0.23 mEq/100 g) (**Figure 3**). Compared with chernozem, calcium content was lower, while magnesium proportion was relatively higher, which may influence soil dispersion and reduced structural stability.

The soil consisted of 41.71% silt and 39.30% clay fractions, indicating a light loam texture (**Figure 4**). Lower organic matter combined with fine particles may increase vulnerability to contamination and enhance mobility of certain heavy metals under changing redox conditions [55].

3.1.4. General interpretation of soil physicochemical status

Overall, all soils were characterized by alkaline reaction and fine-textured composition, but differed significantly in organic matter content and cation-exchange characteristics. These differences are critical for heavy metal retention and electrokinetic transport processes.

Fine-textured soils with high clay and silt content generally enhance adsorption and reduce hydraulic conductivity, which directly affects electromigration and electroosmosis during EK remediation [56]. Meanwhile, soil organic matter can either immobilize metals through complexation or enhance mobility depending on pH and redox conditions [57].

Thus, chernozem exhibited the highest buffering and sorption capacity, followed by light chestnut soil, while light gray soil showed the lowest natural protective capacity against contamination.

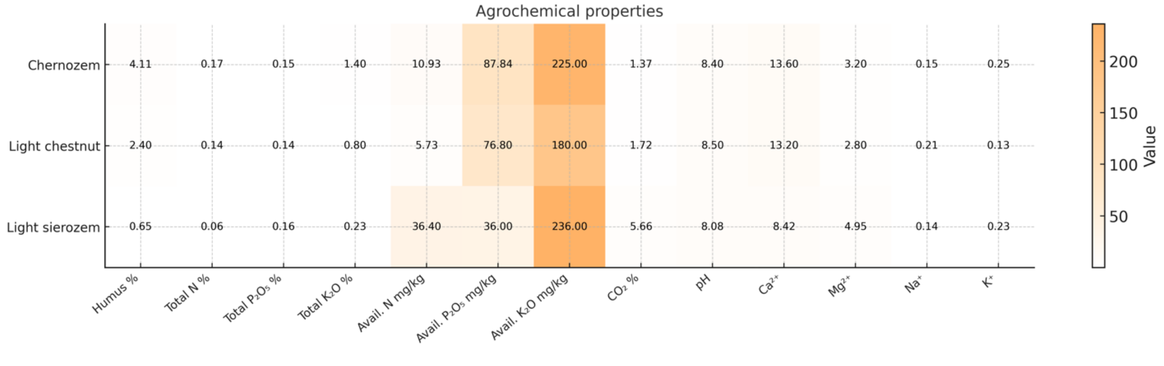

Figure 3. Agrochemical characteristics of soils (0–20 cm).

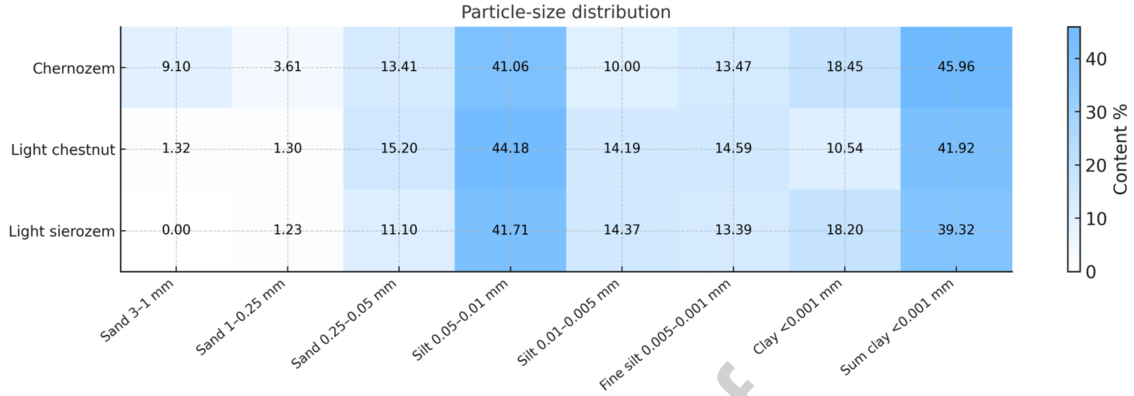

Figure 4. Particle-size distribution of soils (0–20 cm).

3.2. Treatment of heavy metal-contaminated soils

Based on the established baseline physicochemical properties, the effectiveness of electrokinetic (EK) remediation followed by biochar amendment was evaluated for the removal of Cd, Cu, Pb, and Zn from the studied soils. EK remediation is widely recognized as an efficient method for fine-grained soils due to its ability to induce electromigration and electroosmosis, enabling metal transport even in low-permeability matrices [11, 40].

In this study, soils were subjected to EK treatment followed by biochar application at doses of 15, 25, and 35 t/ha. Biochar is known to enhance metal immobilization through adsorption, surface complexation, and pH modification, thereby reducing metal bioavailability after electrokinetic mobilization [58, 59].

The efficiency of remediation was expected to vary significantly depending on soil texture and exchangeable cation composition. Soils with higher clay and organic matter content (chernozem) tend to exhibit stronger metal retention, while low-organic soils (light gray soil) generally show higher metal mobility but lower buffering capacity during electrochemical treatment [60].

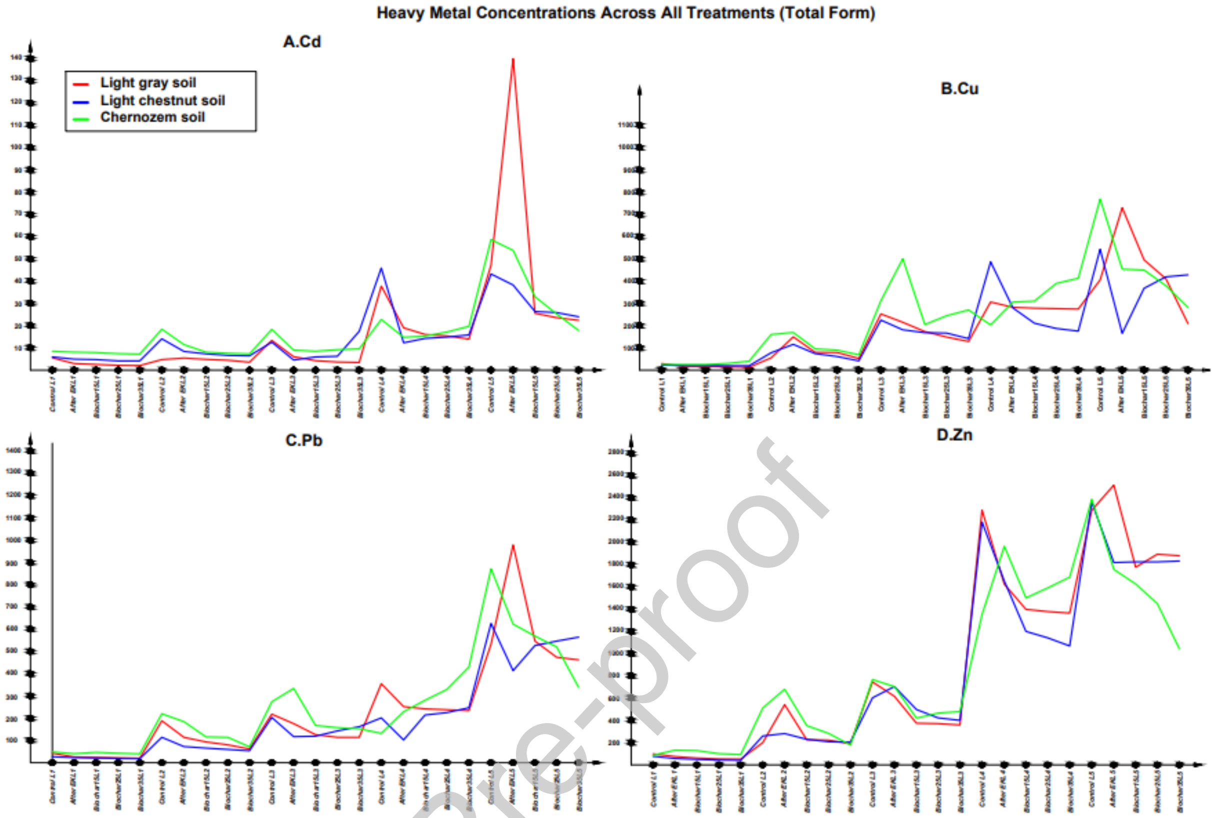

Figure 5. Results of soil heavy metal removal using EK + biochar remediation (total form).

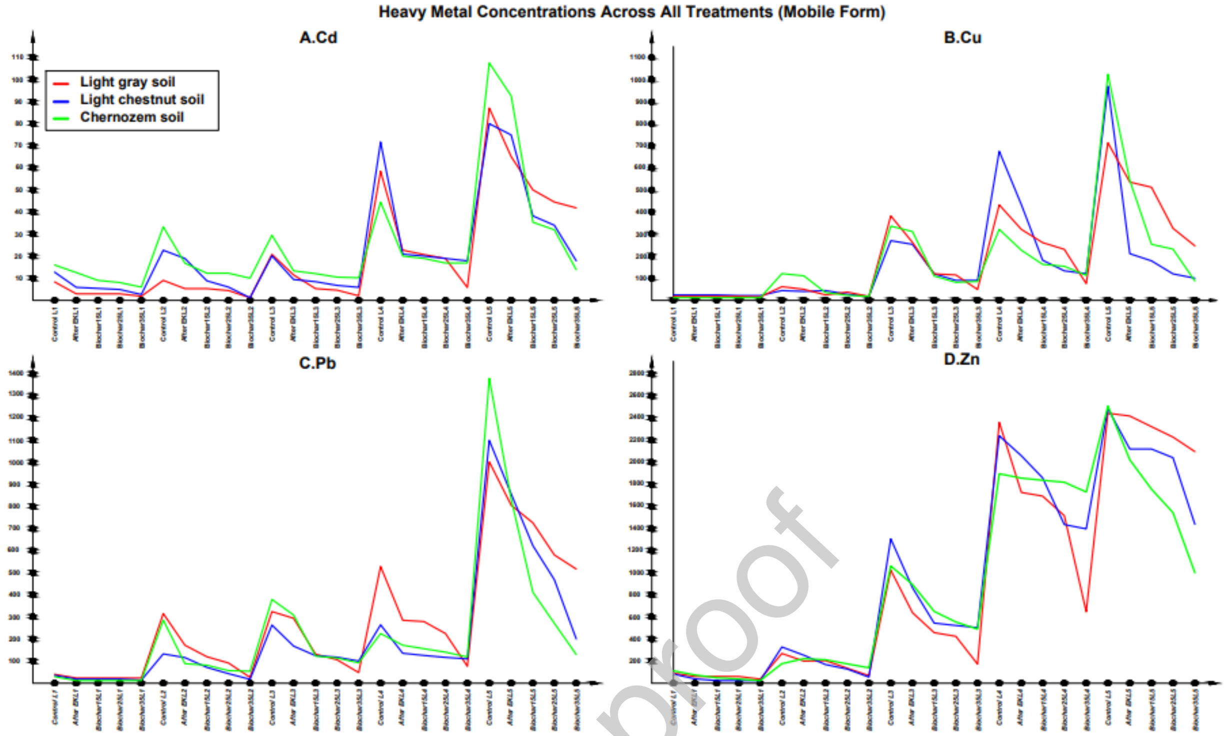

Figure 6. Results of soil heavy metal removal using EK + biochar remediation (mobile form).

The results are consistent with published evidence showing that EK processes promote migration and partial extraction of mobile metal forms, while biochar immobilizes metals through adsorption, precipitation, and cation-exchange mechanisms, reducing their bioavailability and ecological risk [61-64].

3.2.1. Cadmium analysis

Light gray soil: Before remediation, Cd concentrations were 5.52 mg/kg (total) and 9.51 mg/kg (mobile). EK treatment led to a decrease of 60.3% (total) and 59.3% (mobile), indicating strong susceptibility of Cd to electromigration in this soil. Following biochar addition, the efficiency increased in a dose-dependent manner: 15 t/ha: total decrease of 66.1%, mobile decrease of 59.5%, 25 t/ha: total decrease of 74.8%, mobile decrease of 64.5%, 35 t/ha: total decrease of 75.9%, mobile decrease of 80.0% (**Figures 5 and 6**).

The 35 t/ha biochar dose produced the highest reduction, particularly for mobile Cd, consistent with reported immobilization via surface complexation and Cd-carbonate/hydroxide precipitation [16, 65].

Light chestnut soil: Initial Cd concentrations were 5.64 mg/kg (total) and 12.30 mg/kg (mobile). EK reduced these values by 24.6% and 48.6%, respectively. Biochar further enhanced Cd immobilization: 15 t/ha: total decrease of 29.1%, mobile decrease of 54.8%, 25 t/ha: total decrease of 40.8%, mobile decrease of 66.1%, 35 t/ha: total decrease of −40.8%, mobile decrease of 73.7% (**Figures 5 and 6**).

The strongest effect was again observed in the mobile fraction, indicating that biochar predominantly stabilizes bioavailable Cd.

Chernozem soil: Before treatment, Cd amounted to 8.12 mg/kg (total) and 15.20 mg/kg (mobile). EK had limited influence on total Cd (decrease of 2.1%) and a moderate effect on mobile Cd (decrease of 16.0%), reflecting high buffering capacity and organic matter content of chernozem. Biochar amendment significantly improved Cd immobilization:15 t/ha: total decrease of 2.7%, mobile decrease of 36.1%, 25 t/ha: total decrease of 13.1%, mobile decrease of 42.0%, 35 t/ha: total decrease of 13.8%, mobile decrease of 63.7% (**Figures 5 and 6**). This aligns with studies showing that biochar enhances Cd complexation in humus-rich soils [62].

3.2.2. Copper analysis

Light gray soil: Before remediation, Cu concentrations were 15.40 mg/kg (total) and 6.60 mg/kg (mobile). EK reduced total Cu by 73.6% and mobile Cu by 24.7%. Biochar further improved Cu stabilization:15 t/ha: total decrease of 74.0%, mobile decrease of 25.0%, 25 t/ha: total decrease of 77.9%, mobile decrease of 53.3%, 35 t/ha: total decrease of 79.0%, mobile decrease of 61.1%

(**Figures 5 and 6**). The high reduction of mobile Cu at 35 t/ha corresponds with the strong affinity of Cu for oxygen-containing functional groups and mineral phases on biochar surfaces [66].

Light chestnut soil: Initial concentrations: 11.20 mg/kg (total) and 2.89 mg/kg (mobile). EK decreased these by 19.6% and 29.4%. Biochar increased Cu immobilization: 15 t/ha: total decrease of 19.6%, mobile decrease of 30.1%**,** 25 t/ha: total decrease of 21.4%, mobile decrease of 33.9%, 35 t/ha: total decrease of 25.0%, mobile decrease of 50.5% (**Figures 5 and 6**). The mobile fraction showed the strongest response, demonstrating effective Cu stabilization.

Chernozem soil: Initial Cu levels were 14.80 mg/kg (total) and 1.89 mg/kg (mobile). EK slightly affected total Cu decrease of 3.4% but reduced mobile Cu by 46%. Biochar significantly enhanced Cu immobilization:15 t/ha: total decrease of 12.2%, mobile decrease of 63.1%**,** 25 t/ha: total decrease of 23.0%, mobile decrease of 42.0%**,** 35 t/ha: total decrease of 31.1%, mobile decrease of 95.2% (**Figures 5 and 6**). The remarkable 95.2% reduction in mobile Cu at 35 t/ha highlights a synergistic effect between biochar and organic-rich chernozem [63].

3.2.3. Lead analysis

Light gray soil: Before remediation, Pb concentrations were 31.80 mg/kg (total) and 32.60 mg/kg (mobile). EK reduced total Pb by 55.0% and mobile Pb by 54.6%. Biochar further increased immobilization:15 t/ha: total decrease of 58.2%, mobile decrease of 67.5%, 25 t/ha: total decrease of 60.1%, mobile decrease of 70.2%, 35 t/ha: total decrease of 63.8%, mobile decrease of 73.9% (Fig. 5-6). These results correspond with the known strong affinity of Pb for biochar surfaces through precipitation of $PbCO_3$, $Pb_3(PO_4)_2$ and strong surface complexation [67].

Light chestnut soil: Initial Pb: 20.30 mg/kg (total) and 19.60 mg/kg (mobile). EK reduced these fractions by 26.1% and 54.6%. Biochar further enhanced Pb stabilization:15 t/ha: total −29.1%, mobile decrease of 58.7%**,** 25 t/ha: total decrease of 32.0%, mobile decrease of 69.4%**,** 35 t/ha**:** total decrease of 35.5%, mobile decrease of 78.2% (**Figures 5 and 6**). The mobile Pb fraction showed nearly 80% reduction at 35 t/ha, indicating high sorptive capacity.

Chernozem soil: Initial Pb concentrations: 32.30 mg/kg (total) and 26.40 mg/kg (mobile). EK reduced total Pb by 13.9% and mobile Pb by 32.6%. Biochar contributed substantially: 15 t/ha: total decrease of 2.2%, mobile decrease of 47.3%**,** 25 t/ha: total decrease of 9.6%, mobile decrease of 49.2%**,** 35 t/ha: total decrease of 11.1%, mobile decrease of 67.8% (**Figures 5 and 6**). Mobile Pb showed the strongest response, confirming that biochar is highly effective in Pb immobilization in humus-rich soils [67].

3.2.4. Zinc analysis

Light gray soil: Before remediation Zn was 96.10 mg/kg (total) and 67.00 mg/kg (mobile). EK reduced total Zn by 43.1% and mobile Zn by 58.7%. Biochar enhanced Zn immobilization:15 t/ha: total decrease of 60.8%, mobile decrease of 62.4%**,** 25 t/ha: total decrease of 71.2%, mobile decrease of 63.4%**,** 35 t/ha: total decrease of 71.9%, mobile decrease of 76.1% (**Figures 5 and 6**). The high reduction at 35 t/ha aligns with Zn precipitation as carbonates and hydroxides and sorption on biochar surfaces [16, 67].

Light chestnut soil: Initial Zn: 79.90 mg/kg (total) and 67.80 mg/kg (mobile). EK decreased these fractions by 13.8% and 57.4%. Biochar further improved Zn stabilization:15 t/ha: total decrease of 15.8%, mobile decrease of 70.4%**,** 25 t/ha: total decrease of 20.8%, mobile decrease of 83.9%**,** 35 t/ha: total decrease of 21.4%, mobile decrease of 87.9% (**Figures 5 and 6**). Zn immobilization was strongest at 35 t/ha.

Chernozem soil: Initial Zn: 85.10 mg/kg (total) and 76.00 mg/kg (mobile). EK reduced total Zn by 37.8% and mobile Zn by 27.6%. Biochar enhanced this effect:15 t/ha: total decrease of 32.0%, mobile decrease of 33.5%, 25 t/ha: total decrease of 14.6%, mobile decrease of 66.7%, 35 t/ha: total decrease of 21.2%, mobile decrease of 80.0% (**Figures 5 and 6**). Zn immobilization at 35 t/ha reached 80%, consistent with strong Zn sorption onto carbonized surfaces [68].

Assessment of Remediation Efficiency: Across all soil types: EK alone reduced mobile metal forms by 20–60%. EK + biochar reduced mobile forms by 60–95%, depending on metal and soil. Biochar at 35 t/ha consistently provided the highest immobilization efficiency for all metals. These results align closely with global studies demonstrating that combining EK with biochar yields synergistic effects by enhancing both removal (EK) and stabilization (biochar) processes.

3.3. Biological productivity of plants under remediation conditions

This experiment was aimed at assessing the effect of soil remediation using electrokinetic purification (EK) in combination with biochar application on the growth of two plant groups—cereals and legumes - cultivated in three soil types: light gray soil, light chestnut soil, and chernozem. Plant productivity was evaluated using three key indicators: plant number, plant height, and the integral productivity index (PI = number × height).

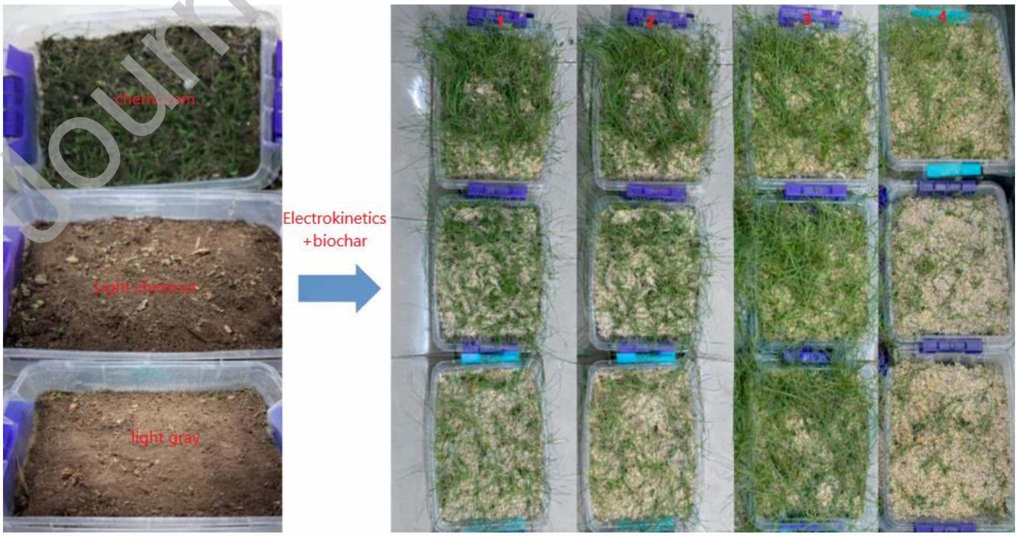

Figure 7. Plants as affected by biochar dose and contamination level.

In the control variant, plant growth in light gray soil was extremely limited due to the toxic effects of heavy metals. Cereals showed 20 plants with zero height, while legumes exhibited only 6 plants with a height of 0.5 cm. Plant growth at contamination levels 4 and 5 in the control variant was completely inhibited (0%), indicating severe phytotoxicity.

Following biochar application, a pronounced improvement in plant growth and development was observed. For cereals, biochar at 15 t/ha increased plant number to 120 with a height of 9.2 cm (PI = 1104). At 25 t/ha, plant number increased to 170 and height to 15.0 cm (PI = 2550), while at 35 t/ha, 220 plants reached a height of 19.9 cm (PI = 4378).

Legumes responded similarly, though with lower absolute values. At 15 t/ha, 40 plants reached 3.1 cm (PI = 124); at 25 t/ha, 56 plants reached 5.0 cm (PI = 280); and at 35 t/ha, 73 plants reached 6.6 cm (PI ≈ 482).

In general, elevating the biochar applications in light gray soil led to a consistent increase in plant count, stature, and productivity index, with the overall productivity measure surging over 4 times in comparison to the control. This suggests a significant decrease in soil toxicity and rejuvenation of biological activity.

Light chestnut soils (**Figures 7 and 8**): In light chestnut soil, the control variant also exhibited minimal plant growth: cereals formed only 24 plants with zero height, while legumes formed 7 plants with a height of 0.5 cm. As in light gray soil, plant growth at contamination levels 4 and 5 was entirely suppressed, confirming the strong toxic impact of heavy metals. Biochar application significantly enhanced growth parameters. For cereals, plant number reached 144 with a height of 9.9 cm at 15 t/ha (PI ≈ 1426), 204 plants with 16.3 cm at 25 t/ha (PI ≈ 3325), and 264 plants with 21.9 cm at 35 t/ha (PI ≈ 5782). For legumes, biochar doses of 15, 25, and 35 t/ha resulted in 48, 68, and 88 plants with heights of 3.3, 5.4, and 7.3 cm, respectively (PI ≈ 158–642). Thus, light chestnut soil demonstrated a clear dose-dependent increase in plant number, height, and PI. The integral productivity index increased by tens to hundreds of times compared to the control, confirming the strong phytostimulating effect of combined EC and biochar remediation.

Chernozem soils (**Figures 7 and 8**) The highest plant growth rates were recorded in chernozem soil. In the control variant, cereals formed 28 plants with zero height, and legumes formed 9 plants with a height of 0.5 cm. Growth at contamination levels 4 and 5 was completely inhibited, indicating total suppression under heavy-metal stress.

Biochar application resulted in a substantial recovery of plant growth. For cereals, 15 t/ha of biochar increased plant number to 168 and height to 10.5 cm (PI = 1764). At 25 t/ha, 238 plants reached 17.6 cm (PI ≈ 4189), while at 35 t/ha, 308 plants reached 23.9 cm (PI ≈ 7361). For legumes, corresponding values were 56 plants with 3.5 cm height at 15 t/ha (PI = 196), 79 plants with 5.9 cm at 25 t/ha (PI ≈ 466), and 102 plants with 8.0 cm at 35 t/ha (PI = 816). Thus, in chernozem soil, all indicators - plant number, height, and integral productivity index - were significantly higher than in light gray and light chestnut soils. This confirms the superior fertility and favorable agrochemical properties of chernozem for remediation and subsequent plant growth.

Across all soil types, increasing biochar doses resulted in a consistent and pronounced increase in plant number, height, and integral productivity index. Cereals dominated in absolute productivity, with PI values reaching several thousand units, particularly in chernozem soil. Legumes also showed substantial improvement compared to the control, with PI values increasing to several hundred units. Among the studied soils, the highest remediation efficiency was observed in chernozem.

These results demonstrate that biochar application following electrokinetic remediation effectively activates plant growth and significantly enhances productivity. According to previous studies, increasing biochar doses gradually stimulates plant growth, although the effect tends to stabilize beyond an optimal threshold. In the present study, the marked improvement observed at a dose of 35 t/ha indicates high sorption capacity of biochar and enhanced biological activity of the soil.

The influence of EK treatment and biochar doses (0, 15, 25, and 35 t/ha) on the productivity of grass and legume phytoremediators was evaluated across the three soil types. Productivity indicators included plant number, plant height, and the integrated Productivity Index (PI = number × height) (**Figures 7 and 8**).

ANOVA results for grasses: The biochar dose (Dose) was the strongest predictor of plant growth: Plant number: $F = 173.46$; $p < 0.0001$; $\eta^2 \approx 0.92$, Plant height: $F = 176.46$; $p < 0.0001$; $\eta^2 \approx 0.92$, PI: $F = 49.11$; $p < 0.0001$; $\eta^2 \approx 0.77$. Soil type and contamination level also influenced growth ($p < 0.0001$), whereas the Soil × Dose interaction was not significant, indicating that positive biochar effects were consistent across all soil types.

The Tukey test revealed significant differences between all dose pairs, demonstrating a clear dose-dependent increase in plant productivity, in agreement with previous studies reporting enhanced biomass production in biochar-treated contaminated soils [69].

Results for legumes: Legumes showed similar trends: Plant height: $F = 202.97$; $p < 0.0001$; $\eta^2 \approx 0.93$. Contamination level significantly influenced performance ($p < 0.0001$), while soil type had weaker effects ($p > 0.07$). Biochar again produced a strong dose-dependent growth increase.

Mean growth trends: Chernozem: plant numbers increased from ~18 (0 t/ha) to ~235 (35 t/ha); heights from 0 to ~18 cm. Light chestnut: ~16 → ~202 plants; height 0 → 16.6 cm. Light gray: ~13 → ~168 plants; height 0 → 15 cm**.** These trends demonstrate that biochar not only reduces metal toxicity but also improves soil structure and nutrient availability, promoting the development of phytoremediators [70].

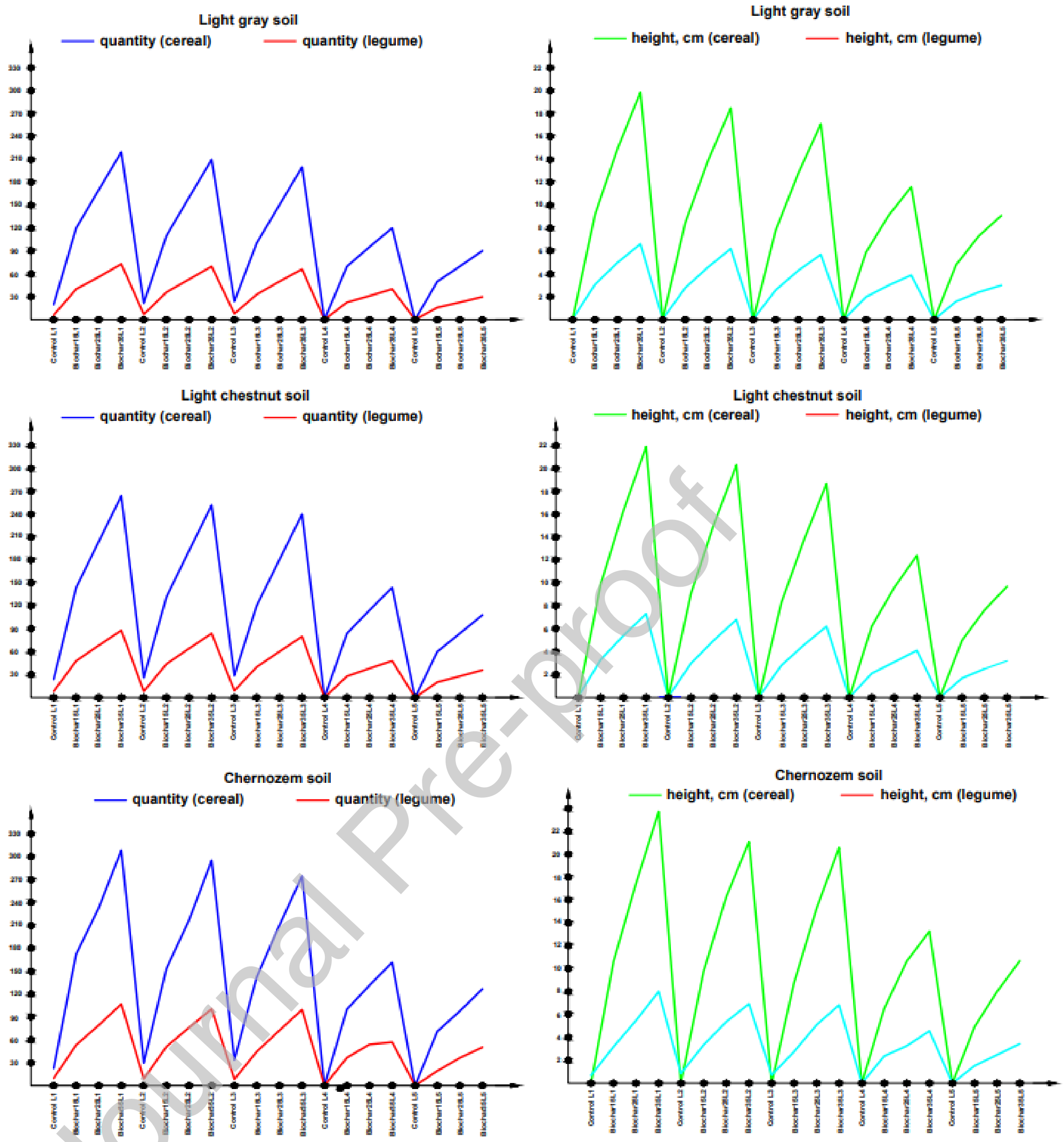

Figure 8. Plants as affected by biochar dose and contamination level.

The statistical analysis demonstrated that biochar dose was the dominant factor controlling phytoremediation productivity, with strong dose-dependent increases observed for both grasses and legumes across all soil types. High F-values ($F > 170$) and large effect sizes ($\eta^2 \approx 0.90$) confirmed the substantial enhancement of plant number, height, and PI following biochar application, while EK treatment acted as a supportive initial remediation step. Chernozem soil showed the highest postremediation productivity, followed by light chestnut and light gray soils. The consistent upward trends across all dose groups indicate that biochar not only mitigates heavy-metal toxicity but also improves soil physical and nutritional status, thereby significantly strengthening the phytoremediation potential of contaminated soils. These results are aligned with previous studies reporting improved plant biomass and stress tolerance in biochar-amended metal-polluted soils [71].

4. Conclusions

This study demonstrated that the combined application of electrokinetic (EK) remediation and biochar amendment substantially reduced both total and bioavailable fractions of Cd, Cu, Pb, and Zn in contaminated chernozem, light chestnut, and light gray soils. The remediation efficiency depended not only on the individual effects of EK treatment and biochar application, but also on their statistically significant interaction.

EK treatment primarily enhanced the mobilization and migration of metal ions through electromigration and electroosmotic transport, increasing the transfer of soluble metal species within the soil matrix. Subsequent biochar application promoted metal stabilization through several complementary mechanisms, including surface adsorption, ion exchange, electrostatic attraction, and complexation with oxygen-containing functional groups. In addition, the alkaline nature and high specific surface area of biochar contributed to increased immobilization of metal ions and reduced their bioavailability. Thus, the observed synergistic effect resulted from the sequential coupling of metal mobilization during EK treatment and physicochemical immobilization after biochar incorporation.

Among the investigated factors, biochar dosage exerted the strongest influence on reducing heavy-metal mobility ($\eta^2 = 0.75$–0.88; $p < 0.001$), while the EK stage also showed a significant independent contribution. The interaction between remediation stage and biochar dose confirmed that the efficiency of metal stabilization increased after EK-induced redistribution of mobile ionic forms. The highest remediation performance was achieved at the biochar application rate of 35 t ha^{-1}, which reduced mobile fractions of Cu and Zn by up to 95% and produced substantial decreases in Cd and Pb mobility across all soil types.

The remediation-induced improvement of soil quality was additionally reflected in biological recovery indicators. Combined treatment significantly increased the productivity of phytoremediation plants, including both grasses and legumes, indicating reduced phytotoxicity and partial restoration of soil ecological functions. Chernozem soil exhibited the greatest restoration potential due to its higher organic matter content and buffering capacity, which enhanced metal retention and supported plant growth after remediation.

Overall, the results establish that the effectiveness of the EK + biochar system is governed by the interaction between electrokinetically induced metal redistribution and subsequent sorption-driven stabilization processes. This integrated mechanism provides a scientifically grounded remediation strategy capable of simultaneously decreasing metal mobility, restoring soil functionality, and improving revegetation potential in heavy-metal-contaminated soils. The findings contribute to a broader understanding of coupled electrochemical–sorption remediation systems and support their application in sustainable rehabilitation of degraded soils in arid and anthropogenically impacted regions.

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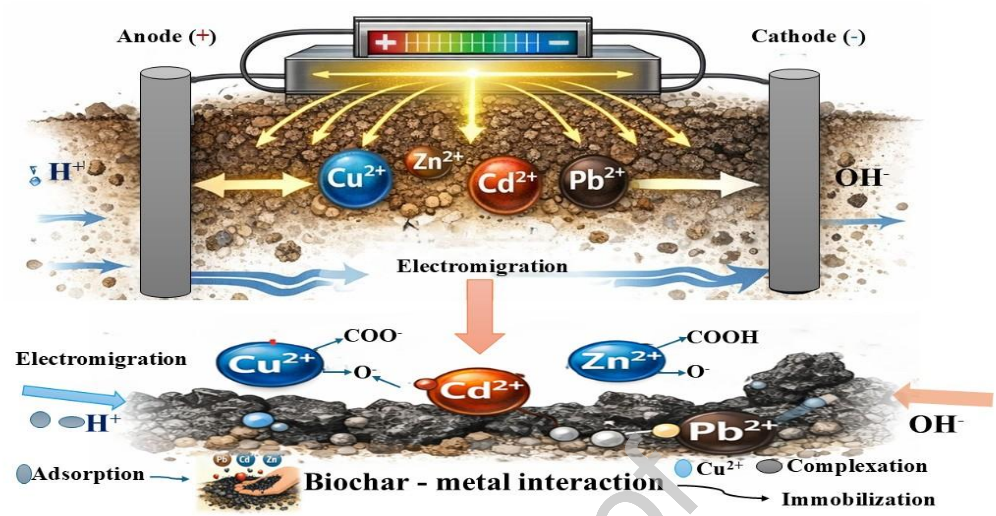
Graphical Abstract



Declaration of interests

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