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Electrokinetics promoted cadmium dissociation, phytoremediation of contaminated plateau red soil and the influence differences between electric field dimensions

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Abstract

Aims The enhanced performance of electrokinetics(EK) on the cadmium (Cd) dissociation, redistribution and phytoremediation of Cd-contaminated plateau red soil has been investigated based on the application of electric field in different dimensions.

Methods After *Sedum plumbizincicola* cutting growth for 30 d, periodic reversal direct-current (DC) electric fields were applied during 150 days for 10.5 cycles.

Results Unlike the uniform distribution change of pH in P1–P9 of the 1D treatment, more soil points (P1–P9) of multidimensional electric fields were exposed to the influence of anode. In electrokinetic–assisted phytoremediation (EKPR), *Sedum plumbizincicola* can alleviate soil acid-alkalization caused by EK, especially the acidification effect of anode under high voltage (10–20 V). Cd speciation and dissociation studies show that EK promotes Cd dissociation into soil pore water, which is conducive to Cd phytoextraction. The periodic reversal DC electric fields enhanced the height more significantly than biomass of *Sedum plumbizincicola* and with inconspicuous among difference regions. Overall, EKPR (voltage of 5–10 V) can promote soil Cd phytoremediation effectively due to the synergistic effect of directly interface action and indirectly influence of electric field to improve the Cd speciation evolution, dissociation, and bioavailability at the soil–water interface. The appropriate electric field arrangement and voltage were EKPR2 and 5 V for *Sedum plumbizincicola*, respectively.

Conclusions EK-induced heavy metals speciation evolution and effective dissociation is one of the important ways to promote the remediation performance, and it is necessary to regulate the arrangement and intensity of electric field to ensure the strengthening effect of EKPR.

Introduction

Soil heavy metal pollution has become a global environmental problem (Song et al., 2022). As far as China is concerned, about 2.786×10^9 m^2 of agricultural soil is contaminated by Cd (Wu et al., 2024; Liu et al., 2015). Remediation of Cd-contaminated soil is extremely urgent. Common contaminated soil remediation techniques include physical techniques, chemical techniques and biological techniques (Song et al., 2022; Wang et al., 2021). Among them, physical techniques such as replacement, vitrification, and solidification have large engineering volume, high cost and energy consumption, while chemical techniques such as oxidation, reduction, and adsorption are easy to produce secondary pollutants and cause large soil disturbance (Hu et al., 2021; Liu et al., 2018; Zhong et al., 2020). Therefore, cost-effective and environmentally friendly bioremediation has been widely studied (Ali et al., 2013).

Phytoremediation is a green and low-cost soil bioremediation technology, which uses the accumulation behavior of heavy metals by plants to absorb or stabilize heavy metals in soil (Wang and Aghajani Delavar, 2024). *Sedum plumbizincicola*, as a Cd hyperaccumulator, has been widely used in the remediation of Cd–contaminated soil due to its fast growth rate and large biomass (Fan et al., 2021;

Zhang et al., 2024). However, the roots of *Sedum plumbizincicola* are shallow, and it can only uptake Cd with high availability within the reach of roots, the remediation efficiency is limited by the accessibility and bioavailability of Cd in the actual contaminated soil (Sarwar et al., 2017). Electrokinetic–assisted phytoremediation (EKPR) involves the application of low–intensity electric fields to stimulate plant growth or promote the absorption of heavy metals by promoting the desorption and short–distance transport of heavy metals (Fan et al., 2021; Ma et al., 2022a). Studies have shown that the growth and heavy metal extraction of *Brassica juncea*, *Sedum plumbizincicola* and *Lolium perenne L.* was promoted in EKPR system. (Cang et al., 2011a; Fan et al., 2021; Ma et al., 2022b; Putra et al., 2013; Sánchez et al., 2020). Current EKPR research focuses on assessing the feasibility of technologies, optimizing parameters, and overcoming adverse effects (Cameselle and Gouveia, 2019; Li et al., 2019; Putra et al., 2013). The selection and application of electric field conditions (electric field type and arrangement, electric field intensity and application time), soil pH control (periodic reversal, alternating electric field and buffer pair) and additives (electrolyte, plant nutrients, soil conditioner, pH regulator and heavy metal chelating agent) had significant effects on the desorption and migration ability, spatial distribution and plant accumulation characteristics of soil heavy metals (Fan et al., 2021; Mao et al., 2015).

Previous studies have shown that the suitable electric field intensity for EKPR is 0.5–4.0 $V \cdot cm^{-1}$. However, the application of direct current (DC) electric field(more than 4.0 $V \cdot cm^{-1}$) for a long time will lead to excessive acidification and alkalization of soil, reduce the number of microorganisms, and cause damage to plants (Luo et al., 2018a, 2019). In order to alleviate the above adverse effects, the application of periodic reversal of DC or AC electric field and pH regulation (adding organic acids, introducing electrolytes with pH buffer regulation function or electrolyte circulation) is currently the main mitigation measures (Han et al., 2021). On the one hand, the introduction of electrolyte makes the soil impregnated (Vocciante et al., 2017), leading to excessive acidification and alkalization, which is not conducive to the growth of most plants. Therefore, the direct electrode connection may be more suitable for plant growth and practical application of EKPR technology. On the other hand, compared with the simulated heavy metal soil, actual contaminated soil under the EKPR system has much lower migration rate and removal rate of heavy metals due to the long-term stability and low bioavailability of heavy metals in the actual soil. Migration and short-distance transport can be adopted to overcome the low accessibility of heavy metals to plants, more importantly, heavy metals are desorbed from the surface of soil particles. The effect of electrokinetic application on the speciation of heavy metals and the distribution of soil-water interface is particularly important for the treatment of actual contaminated soil by EKPR system, which needs further study. Plant biomass and metal uptake can be effectively improved, while the desorption, migration, distribution characteristics and morphological changes of heavy metals under periodically reversed DC or AC electric fields are rarely discussed (Li et al., 2019, 2020).

To our knowledge, EKPR is involved in the complex interactions of chemical and electrochemical processes at the soil-water interface (Serghei et al., 2009; Wang et al., 2021), whether other mechanisms are included is worth exploring to elucidate the mechanism and regulation of the EKPR system. In this study, the actual Cd-contaminated red soil, *Sedum plumizincicola* and Cd were taken as the research

objects. The DC electric field with direct connection and periodic reversal of electrodes was constructed, and the EKPR pot experiment with three electric field dimensions (1 D, 2 D and 3 D) and voltage intensity (5 V, 10 V and 20 V) was set. The main objectives of this study were: (1) to describe soil pH changes; (2) to analyze the spatial distribution, chemical speciation changes and desorption behavior of Cd; (3) to track the growth and Cd accumulation of *Sedum plumbizincicola*. (4) and to explore the relationship between Cd desorption and the influence factors of soil pH and electric field. The mechanism and regulation of the EKPR system were clarified, the reasons for the promotion of Cd accumulation in plants under electrical action were revealed, and the effects of various combinations of electric field configurations were evaluated.

Materials and methods

Test materials

The *Sedum plumbizincicola* seedlings were taken from the farmland soil restoration base of Lanping Jinding lead-zinc mining area, Nanjing Institute of Soil Research, Chinese Academy of Sciences. After one-month cuttage propagation, the well-grown seedlings with a height of about 10 cm were selected for pot experiment. The tested contaminated plateau red soil was taken from Zhehai Town, Huize County, Qujing City, Yunnan Province, China. (N 26°34'7.46", E 103°38'19.72", with an average altitude of 2124 m), which is a typical heavy metal pollution area in Yunnan Province. After air drying, it was passed through a 2 mm aperture sieve for use. Table S1 shows Soil physical and chemical properties.

Experimental design

The pot experiment was carried out in the greenhouse of the eastern campus of Yunnan Agricultural University. A rectangular PVC culture basin (length × width × height = 24 × 15 × 17 cm) (Fig. 1a) was used, and each basin was filled with 0.005 m^3 soil. The electrode materials were graphite electrode plate (length × width × height = 20 × 15 × 0.5 cm) and graphite electrode rod (length: 8 cm, diameter: 3 cm) (Fig. 1a). It is powered by DC power supply (0–100 V, 0–3 A). In order to facilitate the study of soil Cd speciation in different points, the EKPR culture basin was evenly divided into P1–P9 from the plane, and one *Sedum plumbizincicola* was planted in each point (Fig. 1b, Fig. S1). The electrokinetic remediation system (Fig. 1d) of 1D electric field (EK1, EKPR1), 2D electric field (EK2, EKPR2) and 3D electric field (EK3, EKPR3) under three voltages (5 V, 10 V, 20 V) was constructed, and the phytoremediation control system (CK) without electric field was constructed. Each treatment was set up three times replicates with a total of 57 basins (Table S2). In addition, C0 (a no plant and no electric field control) was set. C0 and CK without electric field effect were not partitioned. Water was poured at the amount of about 500 mL per basin every 3 days. Electricity was applied for 8 h (9:00–17:00) every day and the polarity was reversed every 7 days. 14 days is a cycle, 10.5 cycles of electricity, a total of about 150 days. In the last week, porewater, soil and *Sedum plumbizincicola* were collected according to the points. The porewater of Soil was collected in situ by a special RHIZONMOM pore water sampler of the Dutch type (Fig. 1b). The sampling head was composed of white porous hydrophilic filter membrane (length 10 cm, diameter

2.5 mm, average pore size 0.15 um, with reinforcement wire). A negative Luer connector was connected to a syringe for negative pressure collection.

At the same time, according to the soil-water ratio of 1: 30 (W/V), i.e. 10 g of actual contaminated soil passing 100 mesh sieve was added to a 600 mL PVC basin containing 300 mL of different pH (3, 5, 7, 9, 11) solutions to configure the soil suspension, in which the pH was adjusted by 0.1 mol L^{-1} NaOH and HCl solution, graphite electrode plate was arranged in parallel (electrode spacing of 10 cm, with / without 5 V DC voltage). In addition, the Cd cumulative desorption was determined after desorbing Cd continuously 10 times at pH = 3, with an equilibrium time of 1 h under different voltages (0, 5, 10 and 20 V) (Fig. 1c).

Research method

After EK and EKPR experiments, *Sedum plumbizincicola* were collected in different regions, the plant height was measured, and then was harvested dividedly as shoots and roots. The fresh and dry weights of the plant were measured including shoots and roots. After drying and grounding, the Cd content in the plant samples was determined by Atomic Absorbance Spectrometer (AAS–ICE3300, Thermo Fisher Scientific, Germany) after $HClO_4$-HNO_3 digestion (Fan et al., 2021). Soil samples were collected from all points (P1–P9) and naturally dried for being ground through a 20-mesh sieve. Soil pH was determined by water extraction (soil-water ratio 1:2.5 (W/V)) (Kartal et al., 2006), pH meter (pH–3b, Shanghai Rex Instrument Factory, model, China). The total Cd of soil was determined after digested with three acids (HNO_3-HF-$HClO_4$) and diluted with 1.0% HNO_3 solution. The Cd content of porewater was determined after digested by HNO_3-H_2O_2. The modified BCR continuous extraction technique was used to analyze the soil Cd speciation (Huang et al., 2023). Finally, the Cd content was determined by AAS. All chemicals used in the experiments were of analytical grade.

Statistical analysis

Excel was used for statistics and calculation of each measurement index. SPSS 25.0 was used for significant difference analysis ($P < 0.05$) and correlation analysis. Origin 8.0 and Adobe Illustrator 2023 were used for drawing.

The relative migration rate (RMr) was used to characterize the migration of Cd in space. The accumulation characteristics of Cd in plants were expressed by bioconcentration factors (BCF) and translocation factor (TF). The removal efficiency of Cd in soil was expressed by η and the calculation formulas were as follows,

$$RMr = \frac{C_{cz} \text{ or } C_{Iz} - C_{az}}{2 \times C_0} \times 100\%$$

1

$$BCF = \frac{C_{aboveground}}{C_{soil}}$$

2

$$TF = \frac{C_{shoot}}{C_{root}}$$

3

$$\eta = \frac{C_0 - C_e}{C_0} \times 100\%$$

4

In the formulas, (1) C_{cz}, C_{lz} and C_{az} represent the average Cd content (mg·kg^{-1}) in the soil of cathode region, middle region and anode region, respectively. C_0 is the original Cd content (mg·kg^{-1}) in heavy metal contaminated soil, (2) $C_{aboveground}$, C_{soil}, C_{shoot} and C_{root} represent the average Cd content (mg·kg^{-1}), (3) C_e is the soil Cd content after the experiment (mg·kg^{-1}).

Results

Soil pH

In this study, an attempt was made to investigate the relationship between soil pH changes and electric field arrangement and voltage systematically from the perspective of the different points (P1–P9) to lateral (Fig. 2a, b, c) and vertical (Table S3, voltage of 5 V) profile of potting soil. As shown in Fig. 2 (a), both the value and distribution of soil pH in different soil points varied significantly after long-term application of periodic reversal DC electric field (150 d for 10.5 cycles). The degree of soil acidification and alkalization change in points P1–P9 is positively correlated with voltage. The soil pH of point P1–P9 after EK and EKPR treatment presented different distribution and variation rules due to the influence of different electric field dimensions (1D, 2D, 3D). But under the same electric field arrangement, the pH changes were consistent for the same region of P1–P9 with the increase of voltage. For 1D treatments (EK1, EKPR1), the soil pH showed a uniform distribution in the cathode region (P1, P4, P7), the middle region (P2, P5, P8), and the anode region (P3, P6, P9). But the soil pH presented different distribution of cathode region (P5), middle region (P2, P8), and anode region (P1, P3, P4, P6, P7, P9) for 2D and 3D treatments. In this study case, in general, neither EK nor EKPR treatment made the soil pH extremely acidic and alkaline (4.35–7.15 vs. 5.45), due to the soil buffering capacity and periodic change (electrode reversal time was 7 days) of DC electric field direction.

In order to clearly analyze the overall variation ranges of soil pH under different electric field intensities, Fig. 2 (c) integrated and described the variation of soil pH under different treatments according to the above pH zoning rules. From Fig. 2 (b) and Fig. 2 (c), the soil pH change degree of EKPR treatment

effectively reduced, especially the degree of anodic acidification, compare to that of EK treatment. In terms of electric field arrangements, the cathode alkalization effect of the 2D, 3D electric fields more obvious than 1D, whereas the anodic acidification effect was just the opposite. It is not difficult to see from the median, average and quartile values of the box plot for EK treatment (Fig. 2b) that although the largest soil pH appears (7.03–7.15 of 20 V) The synergistic effect of electric field and planting *Sedum plumbizincicola* in EKPR made this acidification phenomenon has been effectively alleviated. Resulting in overall soil pH of EKPR slightly increased in different electric field, especially for low voltage (5 V & 10 V). For this acid red soil (original pH is 5.45), the mean soil pH of CK slightly increased (5.65 vs. 5.45) and there was no obvious change of soil pH in each region. For multi–dimensional electric field, this study investigates the pH change in the vertical direction and under the voltage of 5 V. There is no difference in pH in the surface and bottom soil under the 1D electric field as shown in Table S3, while there is a significant difference in soil pH in the vertical direction of the 2D and 3D electric fields, which followed the order of surface layer > bottom layer.

Soil Cd content, speciation and spatial redistribution characteristics

Figure 3 (a) shows that in EK1, EK2, EK3, the average soil Cd residue ranging from 9.26 to 9.96 $mg{\cdot}kg^{-1}$ were only a slightly difference to the initial value of 9.36 $mg{\cdot}kg^{-1}$. This finding indicates that there is no significant removal effect of single electrodynamics under direct connection conditions, and the Cd content of the surface soil in the multi-dimensional electric field increases. Table S4 reveals that the Cd content in the surface layer (0–6 cm) is greater than that in the bottom layer (6–12 cm) in 2D and 3D electric fields. However, in EKPR, the soil Cd content was lower than the initial value, and the removal rate was 0.4–50.32%. At the same electric field dimension, the Cd removal rate decreased with voltage increase (EKPR1&2, 5–20 V.), while at the same voltage, the removal rate increased with electric field dimension increase (5&10 V, EKPR1-3. Figure 3 (a)). The distribution range and quartiles of Cd content in different treatments varied with the electric field, indicating that the spatial distribution anisotropy of soil Cd may be related to the electric field layout. Figure 3 (b) shows the distribution of Cd at points P1–P9 under various treatments. In contrast to the uniform distribution of Cd in the initial soil, the distribution of Cd in each treatment is uneven and shows regularity. In EK1, the Cd content at points P1, P4, and P7 is higher than that at points P3, P6, and P9, and the disparity becomes more pronounced as the voltage rises. In EK2 and EK3, the Cd content at point P5 is higher than others. Similarly, all the aforementioned points are arranged in the cathode region. Soil Cd migrated from the anode to the middle and cathode (with the cathode region > the anode region) and accumulates in the middle and cathode regions. The relative migration rate (RMr) was adopted to assess the migration degree of Cd and there exist certain differences among the different treatments (from 5 V to 10 V, in the cathode regions: EK1 -0.92–23.98% EK2 5.11–18.02% EK3 7.52–11.52%; in the middle regions: EK1 -3.37–9.64%, EK2 1.70–12.30%, EK3 -0.42–6.68%). The RMr in the cathode region is greater than that in the middle region. Under the same electric field dimension, the RMr in both regions increases with the increase of voltage. Under the same voltage, the RMr in both regions decreases with the increase of the electric field dimension. Compared

with the 1D electric field, the soil Cd distribution was more uneven under the multi-dimensional electric field, and the overall Cd content was higher. In EKPR, the relationship between RMr and voltage as well as the electric field dimension was not obvious, the Cd content at each point in EKPR decreased significantly. A reduction of Cd ranging from 0.94–64% was observed under EKPR treatment compared to the original soil Cd, with the lowest soil Cd content being 3.37 $mg \cdot kg^{-1}$ (EKPR2–5V P3). The accumulation of Cd by *Sedum plumbizincicola* is conspicuous, and the removal law of Cd is 5 V > 10 V > 20 V.

Figure 4 (a) shows that the original soil (C0) Cd mainly exists in the residue state, and the content accounted for as high as 77%. For the Single plant action (CK) residue Cd accounted for 51%, a decrease of approximately 26% compared to C0. The proportions of the remaining three speciations of Cd increased to a certain extent, with acid-extractable Cd increasing by about 14%. In EK, the percentages of both acid-extractable Cd and reduced Cd increased significantly, and the percentage of residue Cd decreased significantly, and the percentage of acid-extractable Cd increased by 19–26% compared to C0, and by 5–12% compared to CK. The effect of a single electric field can increase the content of acid-extractable Cd in the soil, and its performance is better than that of a single plant. Additionally, in EK2 and EK3, the acid-extractable Cd was higher than that in EK1, and the residue Cd decreased as the voltage increased. Figure 4 (b) shows that the Cd content of different speciations in P1–P9 under different treatments. The content law of Cd speciations in most sites was as follows: residue Cd > reducible Cd > acid-extractable Cd > oxidizable Cd. The content of acid-extractable and residue Cd affects the accumulation of Cd in plants. The following results focus on the analysis of these two Cd speciations. In EK1(5–20 V), the acid-extractable Cd content (P1–P9: 5 V ranging from 2.24 to 2.86 $mg \cdot kg^{-1}$, 10 V ranging from 2.42 to 2.95 $mg \cdot kg^{-1}$, 20 V ranging from 2.20 to 3.08 $mg \cdot kg^{-1}$), the higher the voltage, the greater the difference in acid-extractable Cd content at each point. The content of acid-extractable Cd at points P1, P4, and P7 was lower than that at points P3, P6, and P9, while the content of residue Cd was the opposite. Compared with EK1, this pattern is not obvious in EKPR1, but the contents of acid-extractable Cd and residue Cd decrease significantly. In EK2 (5–20 V), the content of acid-extractable Cd changed significantly along with the voltage, and the content of acid-extractable Cd (4.10 $mg \cdot kg^{-1}$) in EK2–20 V P5 reached the maximum. Cd migrated from the anode region (P1, P3, P4, P6, P7, P9) to the cathode region (P5) through electromigration and electrodialysis. In EK3 (5–20 V), the content of residue Cd decreased, and the content of acid extractable Cd increased significantly. Compared with EK and EKPR, the content of acid-extractable Cd was absorbed by plants, and the content of residue Cd, reduced Cd and acid-extractable Cd decreased to a certain extent. Compared with EK2, EK3 and EK1, regarding the content and distribution of Cd speciations, the more the electric field dimension, the greater the difference. Figure 4 (c) shows the Cd content in soil pore water under different treatments. The Cd content in soil pore water increased in electric field. In EK, the soil pore water Cd content rose by 17.78–227.82%, with a maximum of 21.63 $\mu g \cdot L^{-1}$ (EK2–10 V Anode). The variation rule of the soil pore water Cd content with the voltage under the EKPR treatment was not evident. The Cd content of pore water in the anode region was significantly higher than that in the cathode region. However, the

difference in Cd content of pore water in the cathode and anode regions under the three electric fields at 5 V and 20 V was less than at 10 V.

Figure 5 shows the correlation thermograms of soil pH, Total Cd, Cd speciations and Cd in soil pore water. Soil pH is significantly positively correlated with Total Cd ($P < 0.01$) and reducible Cd ($P < 0.01$), and significantly negatively correlated with acid-extracted Cd and soil pore water Cd ($P < 0.05$), Soil Cd content was significantly and positively correlated with residue Cd and reducible Cd ($P<0.01$); acid-extracted Cd was negatively correlated with residue Cd ($P<0.05$) and positively correlated with pore water Cd content ($P<0.05$).

Plant growth and Cd accumulation

The effect of electric field on the growth of *Sedum plumbizincicola* was shown in Fig. 6 (a), the biomass of *Sedum plumbizincicola* was higher than that of CK at 5 V, with an increase of 0.4–17.12%, which promoted the growth of *Sedum plumbizincicola*, while it was lower than that of CK at 20 V, with a decrease of 9.6–39.8%, which inhibited the growth of *Sedum plumbizincicola* obviously. The biomass laws of *Sedum plumbizincicola* were 5 V > 10 V > 20 V under the same electric field, and EKPR2 > EKPR3 > EKPR1 under the same voltage. The height of *Sedum plumbizincicola* was promoted in EKPR, which was increased by 6.2–29.3% than that of CK. The height of *Sedum plumbizincicola* followed the trend: 5 V > 10 V > 20 V. The differences of the biomass and the height of *Sedum plumbizincicola* were not obvious on the different soil regions in EKPR with polarity reversal DC electric field. Figure 6 (b) shows that the Cd content in shoot and root of *Sedum plumbizincicola* increased in EKPR by 4.80–24.29% and 1.77–42.52%, respectively, compared with that of CK (decreased in some regions under some treatments, e.g., EKPR1–20 V), and the law of Cd content in shoot was followed 5 V > 20 V > 10 V in EKPR2 and EKPR3. The Cd transport factor (TF) and Bioconcentration factor (BCF) of *Sedum plumbizincicola* were as shown in Table S5. The maximum TF was 3.06 (EKPR3–20 V, Middle), and the TF increased significantly in EKPR2–5 V and EKPR3–20 V. The Cd BCF of the *Sedum plumbizincicola* were increased by -5.51–108.48% with a maximum of 214.80 (EKPR3–5 V, Cathode). In conclusion, the biomass of the *Sedum plumbizincicola*, did not differ zonally in the electric field, but only had an effect on the overall biomass. However, the accumulation behavior of *Sedum plumbizincicola* to Cd was significantly different in different regions, which showed that cathode > anode.

The overall growth, Cd accumulation, and soil Cd removal rate of *Sedum plumbizincicola* under each treatment are shown in Fig. 6 (c). The design of the electric field had a great influence on the growth and Cd accumulation efficiency of *Sedum plumbizincicola*. Plant growth was promoted in the appropriate voltage range (5 V & 10 V), and plant growth was inhibited by too high voltage (20 V). For the electric field arrangement, the multi-dimensional electric field has denser electric field lines compared with the 1D electric field, which can more easily promote the migration of Cd, thus solving the problem of limited plant root range. Polarity reversal in electrode orientation can prevent extreme changes in soil pH. In this experiment, the growth of *Sedum plumbizincicola*, Cd accumulation and soil Cd removal were unsatisfactory at 20 V. The biomass decreased by 40.31% and the soil Cd removal was only 17.45% in EKPR1–20 V. The growth of *Sedum plumbizincicola*, Cd accumulation and soil Cd removal were highest

under EKPR2–5 V, the biomass increased by 16.59%, Cd accumulation increased by 29.31%, and soil Cd removal was as high as 50.23%.

Effects of electrodynamics on Cd desorption from contaminated soil

The design of the static desorption test with or without electric field of different pH solutions, and try to analyze the reason of Cd desorption from the two aspects of electric field and pH change. The pH changes shown in Fig. 7 (a, b) do not affect the Cd desorption equilibrium time, and the electricity advances the desorption equilibrium to varying degrees. The Cd desorption equilibrium was in the range of 30–60 min without electricity, and the desorption equilibrium time was advanced to 15–60 min under different pH treatments (pH 5–11) after applying a voltage of 5 V. The physicochemical desorption of Cd was more favorable under acidic conditions (pH 3, 5), and was inhibited in alkaline environments. In the range of pH 3–5, the 5 V electric field significantly enhanced the desorption of Cd. Compared with the non-electrochemical system, the desorption content of Cd increased by 36.6–127.8%. In the range of pH 7–11, the effect of electric field on Cd desorption is not obvious. Figure 7 (c) showed that the content of Cd desorbed from the contaminated soil continued to increase with increasing desorption time. In addition, the cumulative desorption content of Cd was positively correlated with the electric field in the voltage range of 0–10 V. Applying 10–20 V voltage increased Cd desorption by 21% compared to that without electric field (0.62 $mg \cdot kg^{-1}$).

Discussion

Soil pH affects heavy metal remediation efficiency by influencing heavy metal speciation, spatial distribution and plant growth. In this study, it was found that the degree of soil acidification and alkalization change in points P1–P9 is positively correlated with voltage, and indicated voltage is the dominant driving factor of soil pH change in EK, EKPR systems (Li et al., 2020). As expected, soil pH showed a polarity trend of cathode region > middle region > anode region, which may be related to the water electrolysis reaction of hydrogen evolution at the anode and oxygen absorption at the cathode (Fan et al., 2021). The cathode alkalization effect of the 2D, 3D electric fields more obvious than 1D. In the cathode region of 2D and 3D electric field caused by more denser electric field lines (Li et al., 2020; Shen et al., 2021). However, the overall acidification effect is even more pronounced. This is mainly attributed to the larger anode area and voltage strength (Cang et al., 2011b; Fan et al., 2021). Others are the migration rate of H^+ in soil is higher than OH^- and the generated OH^- is consumed by the metal ions precipitated under the effect of electrodynamics (Xu et al., 2016). Compared with EKPR and EK, the surface soil pH changed less. These results indicated that *Sedum plumbizincicola* can alleviate the adverse development of soil pH and weaken the acid-base effect of soil simultaneously through regulating secretion behavior (Sun et al., 2019). The plant root is generally distributed in the surface layer of the soil, too low pH will affect plant growth, but lower pH is more favorable for heavy metal desorption (Sánchez et al., 2020). The soil pH of surface layer > bottom layer under multi–dimensional electric field made it possible for plant accumulating more heavy metals by promoting plant growth rather than

inhibiting it, and heavy metals will be desorbed in the bottom layer with lower pH, which will be migrated to the surface layer by controlling the direction of the electric field to be accumulated by the plant (Luo et al., 2018b).

The dissociation, chemical speciation change and spatial migration of heavy metals in soil significantly affect the contact probability between plant roots and metal ions, and plant extraction efficiency of phytoremediation. From perspective of different soil points (P1–P9) and electric field regions (cathode, middle and anode), the Cd content, chemical speciation redistribution characteristics of contaminated red soil have been analyzed after long-term non–closed cycles (150 d for 10.5 cycles) periodic reversal DC EK and EKPR treatment. The EK treatment has no effect on reducing the Cd content in the surface layer of soil. In multi-dimensional electric fields (EK2 & EK3), the Cd content in the surface soil even increases to some extent. This is because the vertical direction of the electric field promotes the migration of Cd^{2+} from the bottom soil to the surface soil through electromigration (Luo et al., 2018a; Putra et al., 2013). In EKPR treatment, the reduction of soil Cd content is mainly attributed to the absorption of Cd by *Sedum plumbizincicola*. Meanwhile, the change of soil Cd content responds to the electric field. There are two reasons for this phenomenon. firstly, the voltage magnitude of the voltage affects the growth of *Sedum plumbizincicola* and thus its Cd uptake. secondly, the multi–dimensional electric field generates denser electric field lines, which enables more heavy metals to be enriched towards the root-accessible area and promotes the uptake of Cd by *Sedum plumbizincicola*. Under EK treatment, the distribution of Cd was uneven, but it showed a law of cathode region > anode region. This is because soil CD migrates from the anode to the intermediate and cathode and accumulates in the intermediate and cathode regions. Relative mobility (RMr) was used to assess the degree of migration of Cd, with RMr in the cathode region being greater than in the intermediate region. The distribution of soil Cd was more uneven under the multidimensional electric field, and the overall Cd content was higher, which was caused by the action of the vertical electric field lines that caused the bottom Cd to migrate to the surface layer, and the surface soil was collected for Cd content determination (Fan et al., In EKPR, the relationship between RMr and voltage and electric field dimensions is not obvious, which may be attributed to plant growth and Cd uptake (Fan et al., 2021).

The removal efficiency of Cd has a significant connection with the Cd speciation. Acid-extractable Cd is easily absorbed, while residue Cd is the most stable and not easily absorbed (Pasricha et al., 2021). The original soil (C0) Cd mainly exists in a residual state, which makes it difficult to remove Cd in actual contaminated soil. However, plant action (CK) may activate Cd and increase the bioavailability of Cd by secreting low molecular weight organic acids, resulting in a decrease in soil Cd content (Sun et al., 2019; Wang et al., 2022). The test results show that the effect of a single electric field can increase the content of acid-extractable Cd in soil. These results indicate that the electric field effect can be used to change the soil physicochemical properties, thus contributing to the release of heavy metal in acid extractable state to the soil environment (Luo et al., 2019; Ma et al., 2022b). On the other hand, the multi-dimensional electric field will cause the electro-migration of Cd from the bottom layer to the surface layer, leading to an increase in the content of acid-extractable Cd (Fan et al., 2021). Acid-extractable Cd in EKPR was reduced compared with that in EK but was significantly higher than CK. The change in the percentage of

each speciation was not obvious, which may be related to the absorption of acid extractable Cd from the soil by *Sedum plumbizincicola* in EKPR. In EK, the higher the voltage, the greater the difference in acid-extractable Cd content at each point. This may be due to the decrease of pH in the anode region of electric field and the increase of acid-extractable Cd content. Compared with EK, this pattern is not obvious in EKPR, but the contents of acid-extractable Cd and residue Cd decrease significantly, which is attributed to the absorption by plants.

Soil Cd desorption directly influences the migration and morphological transformation of Cd in soil, thus affecting the efficiency of plant Cd extraction. The Cd content in soil pore water can directly reflect the desorption of Cd. In EK, the electric field promotes an increase in the Cd content in the soil pore water. The Cd content of soil pore water with voltage was not obvious under EKPR treatment. This may be related to the uptake of Cd by plants. The Cd content of pore water in the anode region was significantly higher than that in the cathode region., which may be caused by both direct and indirect effects of the electric field. Some studies have found that application of a DC electric field can directly promote the desorption of metals from soil particle to soil pore water, and the desorbed heavy metals will undergo electrodialysis and electromigration (Raeisi et al., 2020; Tahmasbian and Safari Sinegani, 2013), which makes the spatial distribution of soil heavy metals different, and Cd is similar to it in this experiment. Furthermore, soil pH affects Cd desorption, while anode precipitation hydrogen acidification facilitates soil Cd desorption, and anode soil pore water Cd content increases. However, the Cd desorbed from the anode migrated to the cathode in the electric field, resulting in the difference of soil pore water Cd content between the two regions. Meanwhile, the results of this study showed that the difference in Cd desorption was more significant in EKPR compared with that in EK the purely electrically mediated system, which may also be related to the regulation of plant inter–root secretions (Wang et al., 2022).

Previous studies have found that low voltage (such as 0.2 $V \cdot cm^{-1}$ for *Lolium perenne.L*. and 1 $V \cdot cm^{-1}$ for *Brassica juncea*) will promote their growth, while high voltage will inhibit their growth (Acosta-Santoyo et al., 2017; Cang et al., 2011b). In this experiment, the growth of *Sedum plumbizincicola* was more sensitive to electric field, and was also promoted under the action of low-voltage electric field (5 V and 10 V), while growth was inhibited under high-voltage electric field (20 V). The low-voltage electric field promotes plant growth for three reasons, first, the electric field in the physicochemical characteristics of the soil and the local concentration of contaminants and nutrients (Cameselle and Gouveia, 2019), second electroosmosis and electromigration increase the opportunity for plant roots to absorb soluble ions (Cang et al., 2012), third, it improves nutrient uptake and enhances plant metabolism by stimulating ion fluxes and ion channels in the root membrane (Mao et al., 2016). The increase in below-ground biomass has increased the potential for plant root uptake and accumulation of heavy metals (Cang et al., 2012).

The main factors affecting the desorption behavior of soil heavy metals include soil pH, Eh and adsorption carrier, etc., among which the role of pH is more obvious, and the applied electric field in electric remediation will also play a role in heavy metal desorption (Cang et al., 2011a; Li et al., 2020; Ma et al., 2022b). The above chapters briefly discussed the pH variation, Cd speciation and desorption, soil

pore water Cd content and distribution under the action of electricity, which focuses on the desorption of Cd, Cd desorption is a single effect of pH and electricity or the result of the two together, which is worth more in-depth discussion. The above test results show that the physicochemical desorption of Cd was more favorable under acidic conditions (pH 3, 5), and was inhibited in alkaline environments, pH is the dominant factor of Cd desorption. But the pH changes do not affect the Cd desorption equilibrium time, and the electricity advances the desorption equilibrium to varying degrees. There are three reasons, first, the increase of pH makes the negative charge on the surface of clay minerals, hydrated oxides and organic matter in soil increase, and the adsorption capacity of Cd^{2+} is strengthened. At the same time, the specific adsorption of Cd^{2+} on the surface of oxides and the stability of soil organic matter-metal complexes increase with the increase of pH. Second, with the increase of pH, the concentration of H^{+}, Fe^{2+}, Al^{3+} and Mg^{2+} in soil solution decreased, and the competitive adsorption with Cd^{2+} decreased, which was more conducive to the adsorption of Cd by soil. Third, the increase of pH is conducive to the speciation of $CdOH^{+}$. The affinity of $CdOH^{+}$ at the soil adsorption site is significantly higher than that of Cd^{2+}, and $CdCO_3$ is also generated (Li et al., 2018; Loganathan et al., 2012; Wang et al., 2020). The desorption of Cd was significantly enhanced by the electric field under acidic conditions (the pH range of 3–5). This result indicates that some direct mechanisms of electrochemical action, such as electrode redox, surface polarization of soil particles and changes in charge density distribution, promote the positive shift of Cd dissociation equilibrium. According to the theory of potential pH and electric field polarization, the dissociation of heavy metals in soil without electric field is mainly chemical dissolution, while chemical dissolution and electrochemical dissolution are combined under the action of electric field. Soil Cd desorption was positively correlated with voltage (0–10 V), indicating that Cd in contaminated soils showed a cumulative desorption effect, which was significantly affected by the strength of the electric field. These results are consistent with previous findings that transferred Cd was much greater than the effective Cd level in EK and EKPR under long-term electrokinetic effects. These results suggest that electrokinetic assistance has a direct electrochemical mechanism to promote the effective and continuous dissociation of Cd at the soil-water interface.

Conclusions

The current studies demonstrated that the variation of soil pH was proportional to the applied voltage (0–20 V) and none extreme acid-alkalization occurred (5.01–7.03 and 4.35–7.15 for EKPR, EK system of 20 V, respectively) due to the alleviate periodic reversal and plant *Sedum plumbizincicola.* After CK, EK, EKPR treatment, the evolution of soil Cd speciation was the residual state transformed to the other speciations with the order of influence being EKPR ≈ EK > CK, especially acid-extraction and reducible state. EK can promote the dissociation of Cd into soil pore water (increased 17.78–227.82% in different region with range of 5–20 V) and the Cd content in soil pore water of EK system was much that EKPR system attribute to the phytoextraction of *Sedum plumbizincicola* efficiently. The electric field will cause Cd to migrate from the anode to the cathode, from the bottom to the surface (2D & 3D). Under the synergistic effect of direct interface action and indirect influence (such as soil pH, EC changes) of electric field, soil Cd can continuously dissociate, migrate, and accumulate in the middle and cathode

region circularly. Compared to biomass, Cd content in shoot and TF, the *Sedum plumbizincicola* height, Cd accumulation in root and BCF increased significantly with the improving of voltage range of 0–10 V (significant inhibitory in 20 V). The suitable electric field arrangement and voltage for promoting the growth and Cd accumulation of *Sedum plumbizincicola* simultaneously were 2D, 5 V respectively. And the biomass, Cd accumulation and soil Cd removal rate were 18.21g·pot^{-1}, 960.62 mg·kg^{-1}, 50.23%, and improved by 16.59%, 29.31%, and 28.76% compare with CK, respectively. In summary, electrokinetics can promote *Sedum plumbizincicola* growth, soil Cd accumulation and removal in EKPR system by elevated the evolution of Cd speciation and spatial distribution, soil Cd dissociation, and bioavailability.

Abbreviations

Cd Cadmium
EK Electrokinetics
EKPR Electrokinetic
assisted phytoremediation
RMr Relative migration rate
BCF Bioconcentration factors
TF Translocation factor
removal efficiency of cadmium.

Declarations

Funding statement

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Data availability statement

, Data are available within the manuscript.

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Figures

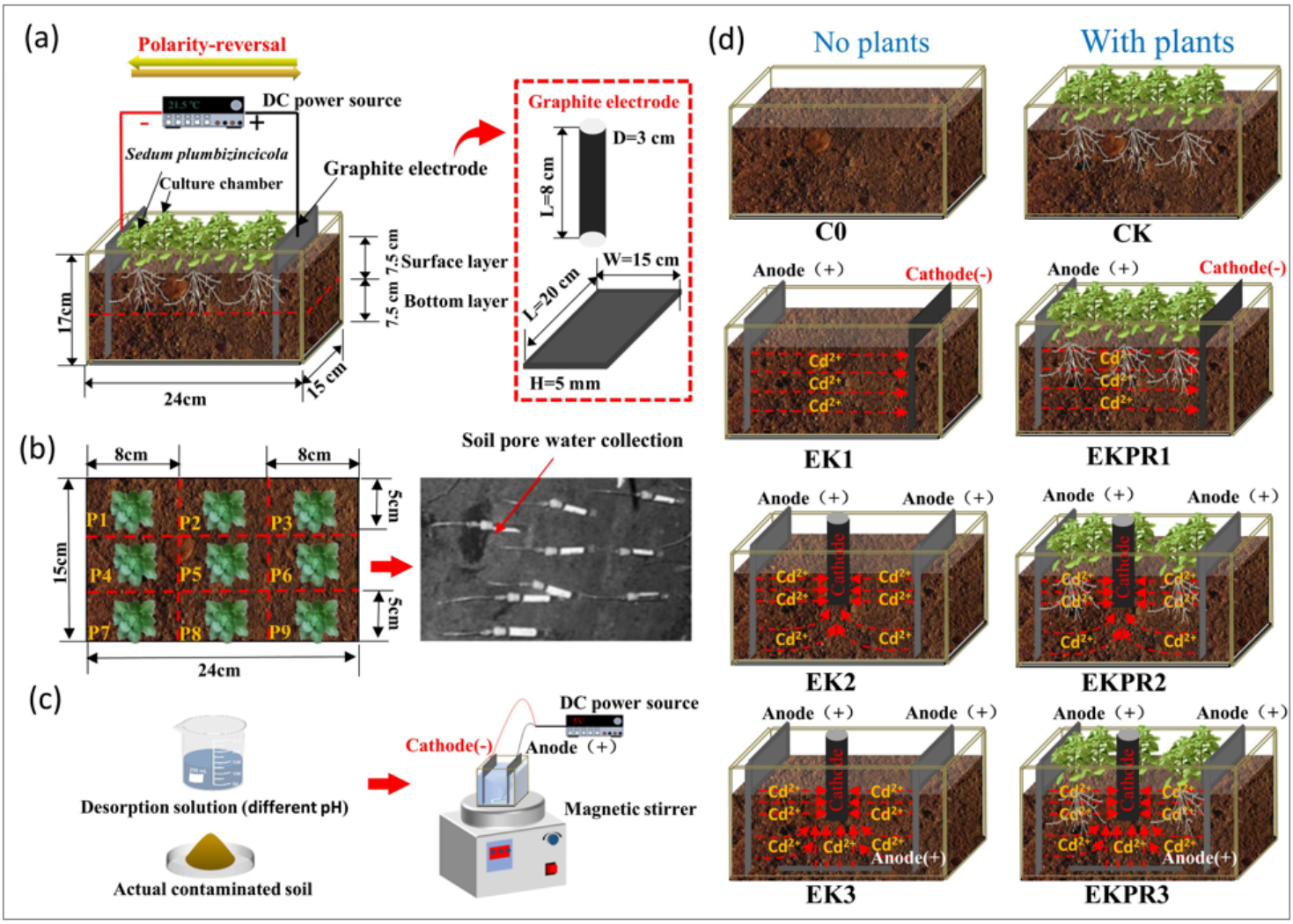


Figure 1

Diagram of EKPR system. (a) Diagram of EKPR device and graphite electrode, (b) P1–P9 points and soil pore water collection diagram, (c) Static desorption test, (d) control and three kinds of electric field layout diagram.

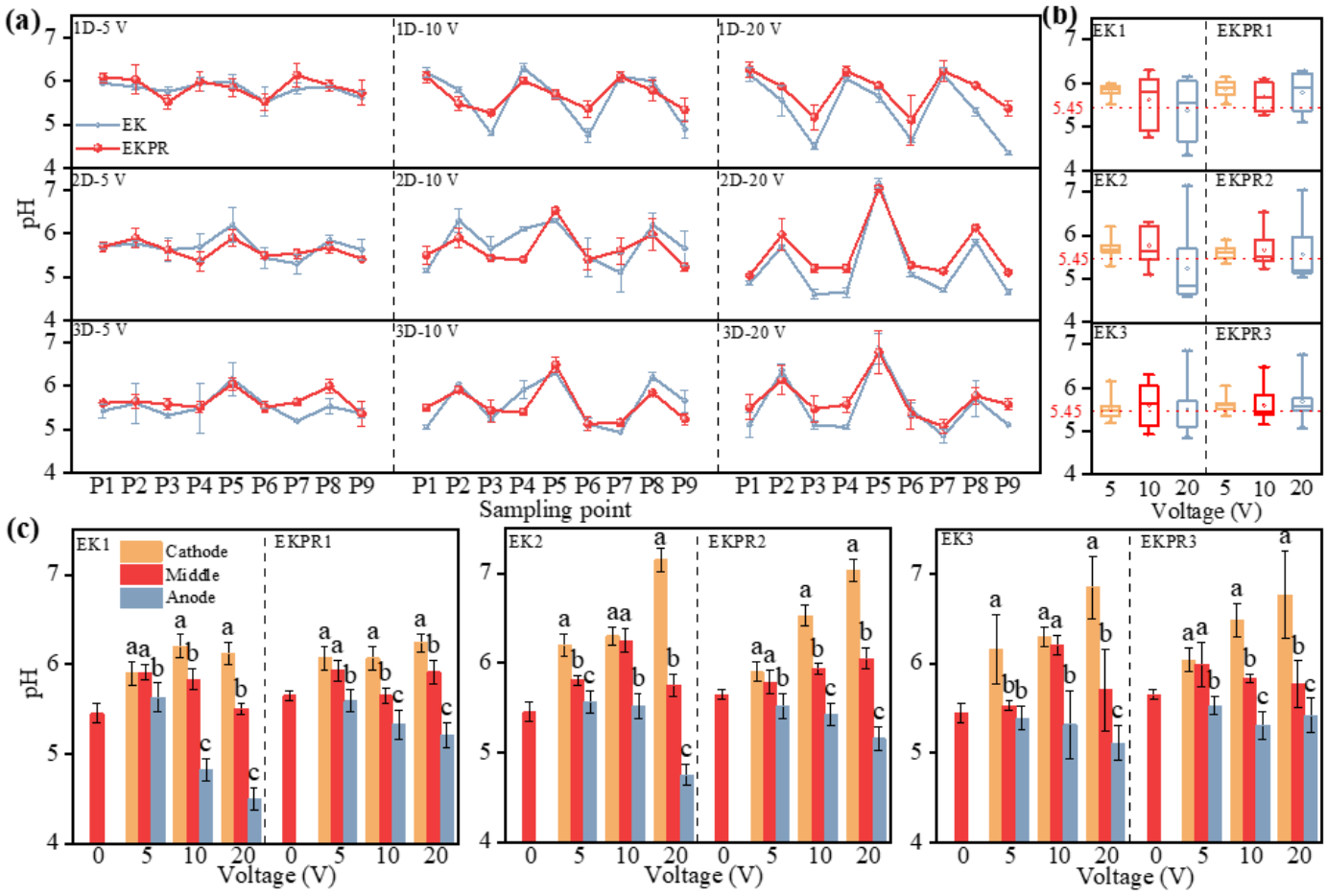


Figure 2

Soil pH under different electric fields. (a) pH of P1–P9 points, (b) Box plot of soil pH, (c) Overall variation ranges of soil pH described in cathode, middle and anode regions after integrated the region of P1–P9 with same rules (Bars represent means ± S.E. of three independent replicates. Those not sharing the same letters are significantly different at $P < 0.05$, as determined by Duncan's multiple range tests).

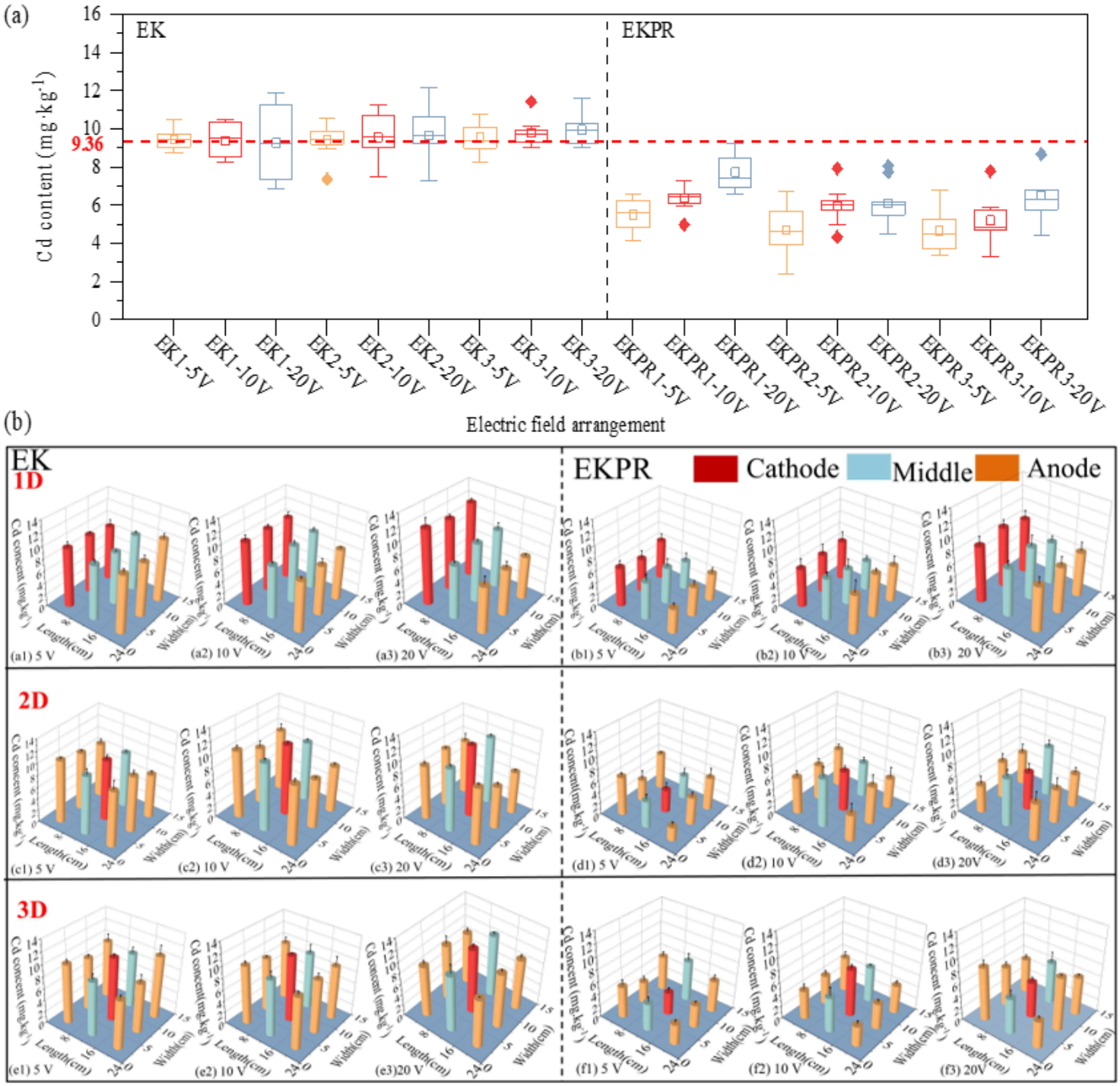


Figure 3

Soil Cd content under different electric fields. (a) Box plot of Cd content under different treatments, (b) Distribution of soil Cd content in P1–P9 points under different treatments

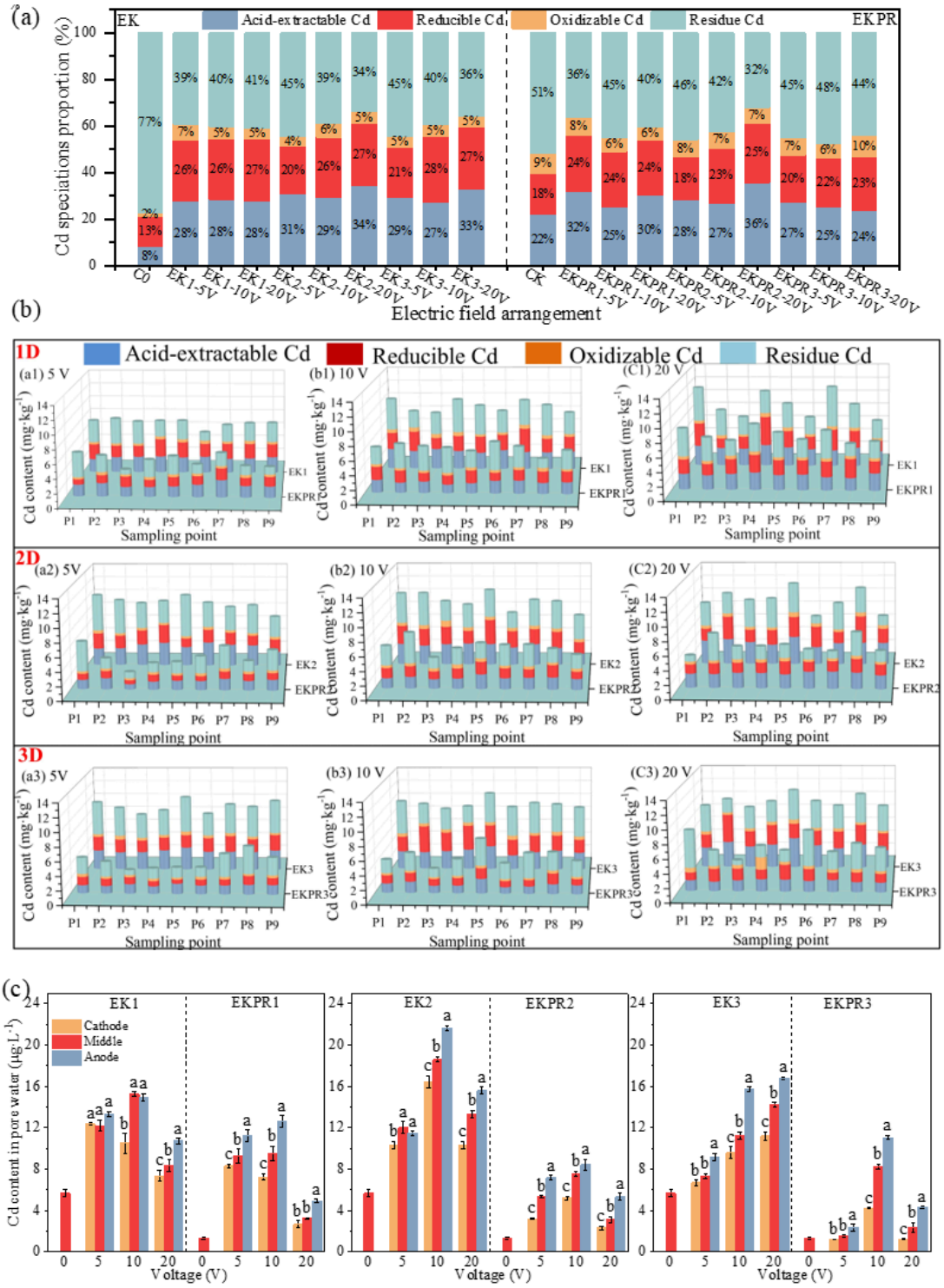


Figure 4

(a) The proportion of Cd speciations under different treatments (the average proportion of four speciations of Cd content in each point), (b) The content of Cd speciations in soil of each point under different treatments, (b) Cd content of porewater in the cathode–anode regions under different treatments. (Bars represent means ± S.E. of three independent replicates. Those not sharing the same letters are significantly different at $P < 0.05$, as determined by Duncan's multiple range tests)

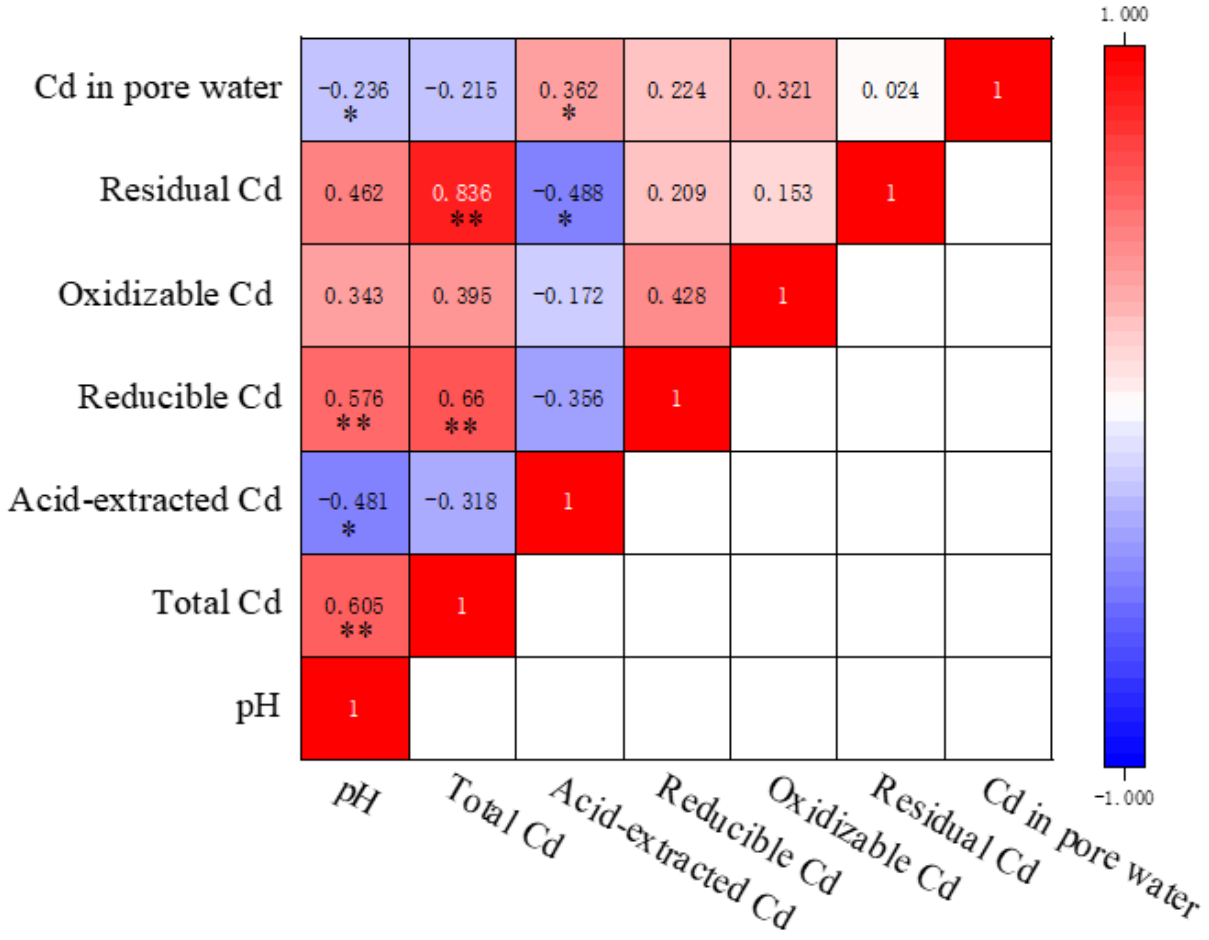


Figure 5

Correlation between soil pH, Total Cd, Cd speciations and Cd in soil pore water. (*, significant correlation at the 0.05 level (bilateral), **, significant correlation at the 0.01 level (bilateral))

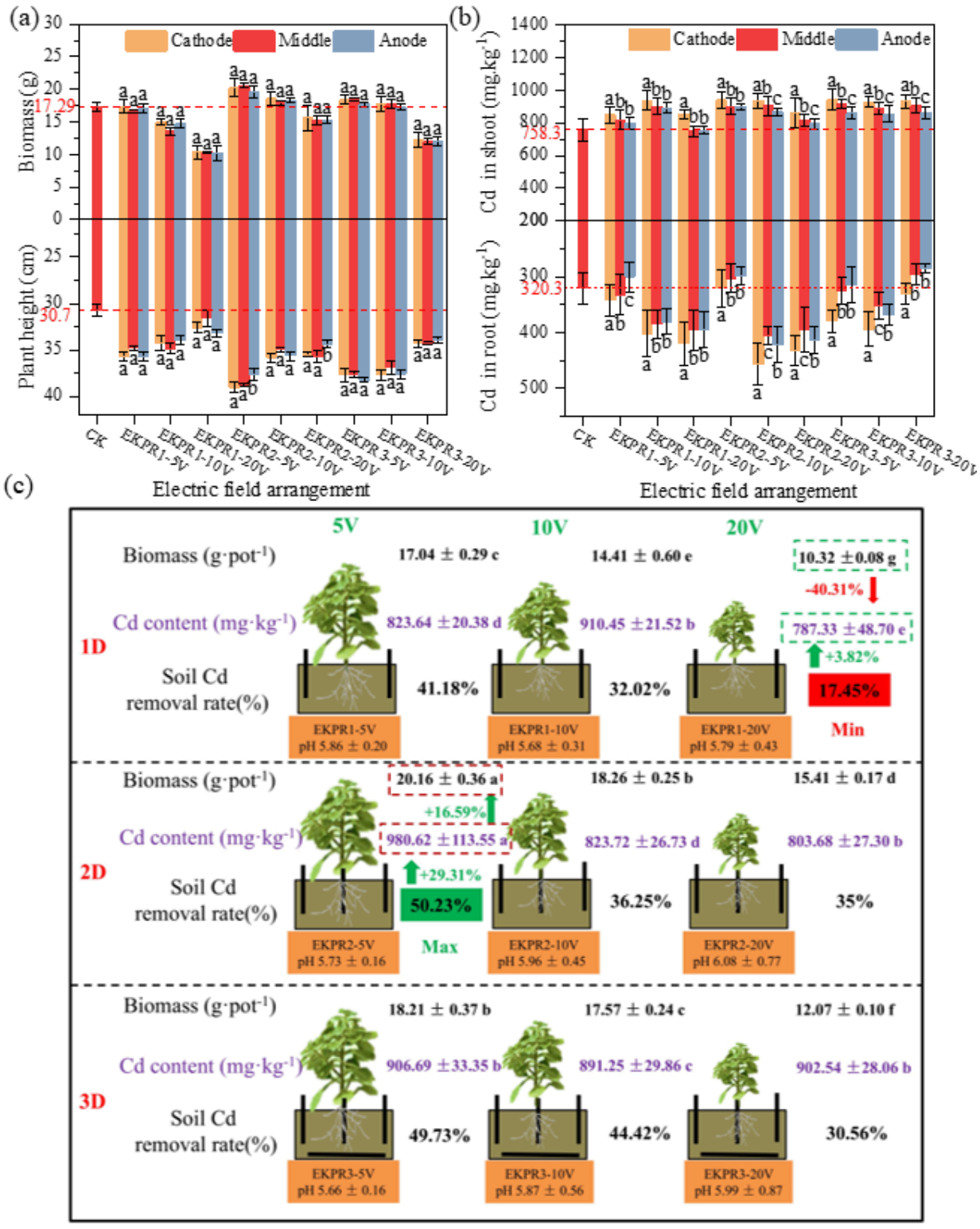


Figure 6

(a) The growth situation of *Sedum plumbizincicola*in the cathode-anode regions, (b) The Cd uptake of *Sedum plumbizincicola*in the cathode-anode regions, (c) The growth, Cd accumulation and Cd removal efficiency of the overall *Sedum plumbizincicola* under different treatments. (Bars represent means ± S.E. of three independent replicates. Those not sharing the same letters are significantly different at $P < 0.05$, as determined by Duncan's multiple range tests).

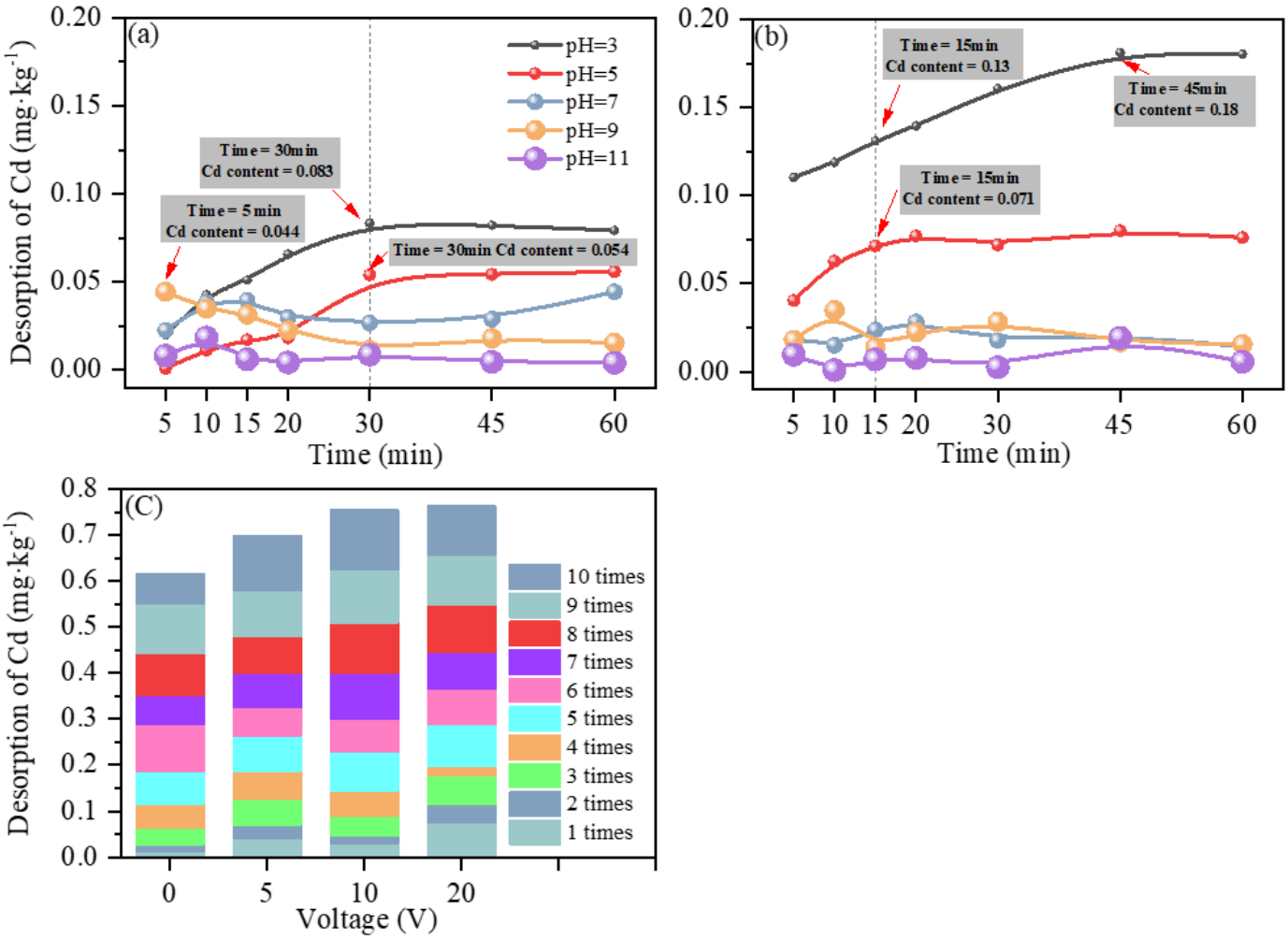


Figure 7

Effect of electrochemistry on the desorption of Cd from contaminated soil under different pH conditions. ((a) without electric field, (b) 5 V applied, (c) pH = 3, and 10 consecutive desorptions under different voltage conditions)

Supplementary Files

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