Journal of Environmental & Analytical Toxicology
ISSN: 2161-0525
Mao, et al., J Environ Anal Toxicol 2018, 8:5
DOI: 10.4172/2161-0525.1000586
Review Article Open Access

Mechanisms and Influencing Factors of Electro-Kinetic Enhanced Phytoextraction for the Recovery of Metal-Polluted Soils

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Received: September 26, 2018; **Accepted:** October 02, 2018; **Published:** October 28, 2018

Abstract

Soil contamination with various toxic metals has become a serious global concerned issue due to the potential risks to the environmental ecology and human health. So far, numerous *in-situ* and *ex-situ* methods have been developed and electro-kinetic (EK) enhanced phytoremediation is widely accepted as a sustainable alternative for the decontamination. The integrated technique contains the application of a low intensity electric field adjoined to growing plants in contaminated soil. So far, many efforts have been made while there is a clear lack of this technology in both laboratorial and field applications. Limitations such as low phytoavailaibility of target metals, unsatisfactory metal's tolerance and translocation ability of plants and undesirable soil pH environment might the main reasons that responsible for the low remediation efficiency. In order to figure out the current focusing phenomena, the possible mechanisms of phytoextraction, EKR and EKR-enhanced phytoextraction were discussed, respectively. In addition, influencing factors such as metal speciation, plant selection and electro-kinetic parameters were also discussed. The contents summarized in the present paper are believed to be an useful guideline for further investigation and optimization of EK-enhanced phytoextraction.

Keywords: Metal-polluted soils; Phytoextraction; Electro-kinetic remediation; Phytoavailability; Electro-kinetic parameters

Introduction

Due to the potential risks to the environmental ecology and human health, heavy metal contamination of soils has become a serious global concerned issue [1]. Generally, the background values of heavy metals such as Cr, Mn, Co, Ni, Cu, Zn, Cd, Sn and Pb are low in soils [2-4]. However, as rapid industrialization and urbanization, their contents in pedosphere, biosphere and hydrosphere have been largely increased in recent years. According to the reports, many countries such as Japan, Indonesia and China are suffering from the heavy metal pollution, which are mainly originated from anthropogenic activities like industrial activities, agricultural productions and mining. Among the above-mentioned pollution sources, agricultural production is believed to be an important contributor since various species of metals have been detected in agro-chemicals such as fertilizers, manures and pesticides, etc. [5]. For instance, superphosphate fertilizers, sludge and lime were found to possess certain concentration of Cd, As, Ni and Cr [6]. In addition, fungicides and insecticides were also reported to contain certain levels of Pb, Cd, As and Zn [7,8]. As a result, phenomena of higher metal accumulation in soils and plants are usually obtained due to the long-term simultaneous and excessive applications of such agrochemicals in arable lands [9,10]. Several researchers have indicated that massive utilization of phosphate fertilizer could result in the content elevation of toxic Cd, As, Cr and Pb in cultivated soils [11]. Moreover, the widespread usage of nitrogen fertilizers which are enriched of Cd rise great risks to soil quality and food safety, even though the concentration of Cd in fertilizers is within the safety threshold [12]. Furthermore, the frequent irrigation with sewage also plays as a vital approach for metals of Pb, Cd, As and Zn to enter the soil environment [13-15].

In order to remediate the metal-contaminated sites, numerous *in-situ* and *ex-situ* methods involving soil replacement, thermal desorption, chemical leaching and fixation, vitrification and biological immobilization have been proposed and implemented based on the efforts spent in the last two decades [16]. However, there is a clear limitation on their widespread applications in field-scale due to the low remediation efficiencies and undesirable site conditions. Therefore, the development of innovative technology with characteristics of cost-effectiveness and environmental-friendliness becomes an urgent issue.

Phytoextraction is widely accepted as a sustainable alternative for decontaminating the toxic heavy metals in soils by using different plants that capable for metal accumulation [17-35]. It was first proposed by Chaney et al. in 1983 and suitable for removing metals, radionuclides, inorganic salts and organic matters from matrix via mechanism of phytoextraction [35]. Even through phytoextraction is a novel, solar-driven efficient and easy applicable strategy, some existing restrictions still hinder its practical utilizations [36,37]. On one hand, it is difficult to find hyperaccumulators that hold favorable accumulation abilities of multiple heavy metals. On the other hand, to ensure the phytoavailability of metals in porous matrix is not easy [38].

For those reasons, various enhancements have been presented to associate phytoextraction and EKR was proved to have preferable improving performance. In brief, the EKR involves introducing a direct low-intensity electric field to mobilize target pollutants through the porous medium. The main transport mechanisms for EKR process to mobilize the metal ions include electromigration, electroosmosis and electrophoresis. As the driving force, the electric potential is beneficial for the transportation of ions toward plant's rhizosphere, which might significantly improve the phtoextraction efficiency. Meanwhile, the

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acidification effect induced by water electrolysis favors the desorption of metals from soil particles, thus largely increases metal's phytoavailability.

The integrated technique of phytoextraction and EKR is promising and worth studying in future due to its huge potential on metal decontamination. In order for better understanding of this technology, the paper described the mechanisms of phytoextraction, EKR and EKR-enhanced phytoextraction, respectively. Then, possible influencing factors which include metal speciation, plant selection and electro-kinetic parameters were also summarized and discussed for further optimization of this technique.

Mechanisms of EK-enhanced phytoextraction

Phytoextraction

Phytoremediation refers to the utilization of plants for removing, stabilizing or degrading the pollutants in environment. That is to say, the various contaminants could be degraded by plants' metabolisms, accumulated in plants' underground and aerial tissues or transformed to other species via rhizofiltration, phytoextraction, phytostabilization, phytotransformation, phytostimulation and phytovolatilization [27,39-42].

Among the above-mentioned mechanisms, phytostabilization and phytoextraction are the most popular approaches for the removal of heavy metals from soils, which aim at controlling the ecological risks and recovering market valuable elements (Ni, Tl, Au) in the meantime. In terms of phytostabilization, it can significantly immobilize heavy metals through effects of root sorption, complexation, precipitation and metal (valence) reduction, thus preventing metal migration to groundwater and food chain [44-46]. As reported, with plantation of proper plants, the hazardous metals could be converted to less toxic fractions due to the function of certain redox enzymes, which largely reduced their mobility and bioavailablity in the environment [43]. In addition, the phytostabilization can also alleviate the accumulation of heavy metals in biota that further decline metals' relevant risks and damages [47]. Regarding to phytoextraction, it is divided into induced phytoextraction and continuous phytoextraction, which can remove metals from the contaminated land by extracting them to the harvestable parts of plant [48]. Generally, induced phytoextraction refers to using plants with remarkable biomass while continuous phytoextraction refers to applying hyperaccumulators with preferable extraction ability [49].

The purpose of phytoremediation is transferring or concentrating heavy metals in roots and above-ground parts of plant at high levels. However, despite the benefits and capacities, there is a obvious shortage of its applications in both laboratory and field scale. Effects such as the speciation of target metals as well as the growth of plants might be the main limitations for the widespread usage of this technique. On one hand, the lower bioavailability of investigated elements would restrict their migration to plants' rhizosphere, thus further hinder the clean-up process [50]. On the other hand, the undesirable plants' growing conditions (e.g., climates, geology, altitude and temperatures) and slower plants' growing and biomass production rate would also limit the performance of phytoremediation [51-55].

Electro-kinetic remediation

Among the various technologies, the EKR is widely accepted as the feasible alternative *in-situ* technique for the enhancement of phytoremediation on soil metals decontamination. Under the constant voltage gradient, metals in porous medium are mobilized towards the opposite electrode direction through mechanisms of electromigration, electroosmosis and electrophoresis (Figure 1) [56].

As so far, many studies have proved that electromigration is the dominant mechanism for cation's transport and their behaviors in soil matrix are significantly influenced by the coupling effects of chemical and geochemical reactions induced by electric field [57-60]. For instance, H^+ and OH^- generated via water electrolysis result in the acidification and alkalization in the anode and cathode regions, respectively. Soil acidic environment is preferable for desorption of toxic metals while alkalic environment can cause species precipitation [61]. As a result, the mobilization of soluble ions are restricted by the effects of adsorption on soil particles and clogging of soil pores in alkaline condition [62].

It seems that the approving performance of EKR on metal recovery can hardly achieved without control on soil pH. For that reason, many researchers have focused on the soil amendments (e.g., chelate, organic acid, etc.) and discovered their great superiority on soil pH management [63-66]. However, significant drawbacks in the damage of soil fertility and secondary pollution were also obviously observed [67-70]. Moreover, due to the relative high expense, promotion of such reagents in large-scale practice was usually difficult to implement.

Recently, great concerns have been paid to the EKR operating parameters (e.g., electrode, materials, electrical field intensity, mode of voltage application and electrodes configurations) in order to improve the efficiency of EKR-enhanced phytoremediation without additional hazard and exorbitant cost. It was reported that, with optimization of aforementioned parameters, the required time for the remediation could be highly diminished and secondary pollution during the process could be largely avoided. Thus, more detailed studies on appropriate EKR designation for different scale requirements becomes an urgent priority.

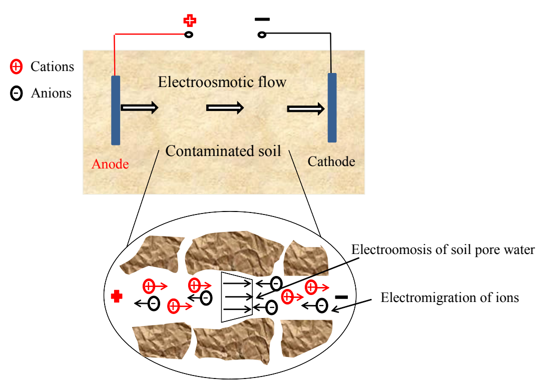

Figure 1: Main mechanisms during an electro-kinetic remediation.

EK-enhanced phytoextraction

In order to avoid the unnecessary remediation time and efforts spent on the soil metal decontamination, the combination of EKR with

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phytoremediation was proposed and tested in different scales [71]. Generally, the integrated technique contains the application of a low intensity electric field adjoined to growing plants in contaminated soil (Figure 2).

Recent researches showed that, the success of EK-enhanced phytoextraction is highly dependent on the bioavailability of metals in soil, meanwhile, the tolerance and translocation ability of plants to target metals [72-74]. Before metal(loid)s enter into plant systems from soil solution, they must be transported to root surface. However, most metals are mainly presented in solid phases such as carbonate-bounded, organic-bounded and iron/manganese oxide-bounded fractions, which can hardly move [75]. Therefore, how to break metals' equilibrium and transfer them into the aqueous fractions becomes a key issue. In addition, selection of plants that hold high translocation factor (TF) of target elements also play an important role in improving metals' extraction efficiency [76].

In conclusion, understanding bioaccumulation and transport of metals by plants under EKR is essential to further recovery of polluted-sites. That is to say, the detailed mechanisms of metal decontamination with EK-enhanced phytoextraction, especially the influencing factors that responsible for the remediation performance, should be better clarified and comprehended.

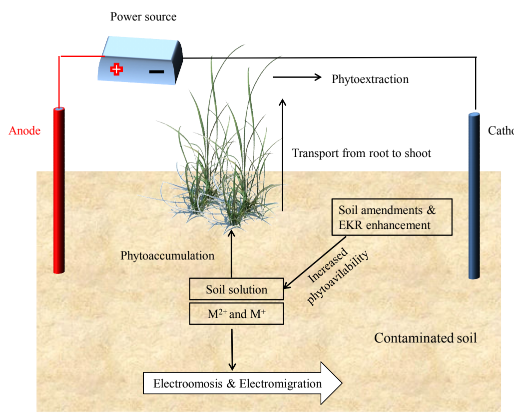

Figure 2: Schematic diagram of electro-kinetic enhanced phytoremediation.

Influencing factors of electro-kinetic enhanced phytoremediation

Metal's speciation and phytoavailability: It is well known that metals are distributed in various chemical forms in soils. Nevertheless, it is also suggested that only water and acid soluble fractions of metals are available for electromigration or plants' uptake. These fractions reflect the phytoavailability of trace metals, which is critical for evaluating the efficiency of EK-enhanced phytoextraction.

Basically, it is hard to estimate metals' speciation due to their complicated chemical activities in both environmental and biological systems [77]. As reported, metal's distribution in soil matrix is determined not only by their concentration but also environmental factors such as water availability and soil properties [78]. Hence, sequential extraction procedures are applied to selectively extract metals with a series of reagents, which aim at characterizing the partitioning of trace metals [79]. A seven-step sequential extraction method was first introduced to analyze metal's speciation by Tessier et al. in 1979 [80]. Then, for the purpose of saving time and avoiding standard deviation, many researches simplified the above analyzing method to a four-step procedure via applying extraction reagents with higher selectivity [81]. Currently, a modified three-step sequential extraction procedure proposed by Community Bureau Reference (BCR) is most popular and widely used associated with diffusive gradients thin-films technology [82].

Based on speciation analysis, soil metals are generally divided into four fractions that named exchangeable, iron/manganese oxide, organic-bounded and residual fraction, respectively [83]. According to phytoavailability, they are further divided into three categories that include readily bioavailable (Cd, Ni, Zn, As, Se, Cu), moderately bioavailable (Co, Mn, Fe) and least bioavailable (Pb, Cr, U). As mentioned before, increasing the phytoavailability of soil metals is critical for improving the efficiency of EK-enhanced phytoextraction. For that reason, enhancement that derived from plant itself and external strategies have been investigated and proposed [84]. For instance, the secretion of H^+ ions by roots will compete the binding sites on soil particles, which lead to the release of metal cations via ion exchange. Moreover, activities of rhizospheric microbes will increase the generation of aqueous complexes, which are also beneficial for metal dissolution through the chelating effect [85]. Referring to external strategy, the purpose of EKR is to manage the pH of treated matrix in an acidic range, which enable to keep more metals in labile fractions. In addition, the application of chelating agents such as EDTA, ammonium sulfate, critic acid and elemental sulfur et al. are usually associated with the remediation process for further reinforcement [86]. As a result, more metals are demobilized around rhizosphere and ready for phytoextraction with the integrated enhancements above.

Plant selection: Generally, plant that ideal for EK-enhanced phytoextraction should satisfy the requirements such as high growth rate, easy to cultivate and harvest, high resistance to pathogens and pests, more above-ground biomass, highly developed root system, tolerance to targeted metals at high content, remarkable metals accumulation and translocation ability, and good adaptability to environmental and climatic changes [87-89]. It was reported that most plants metal decontamination have shallow roots, small aerial biomass and are specific to one element. Hence, great interest have been raised to explore the hyperaccumulators which possess characterizations of fast growing as well as desirable metal accumulation capacity. Hyperaccumulators are plants that hypertolerant to a certain range of metals accumulated in shoots. That is to say, the concentration of metals in their above-ground tissues is usually 100-1000 fold larger than those presented in the soil or in nearby non-accumulators [90]. Currently, there is no scientific standard to define hyperaccumulaor. Thus, hyperaccumulator selection for EK-enhanced phytoextraction can still be in challenge.

The concept of "hyperaccumulator" was first proposed by Brooks et al. and further described by Reeves as plants which capable to accumulate more than 1000 mg kg^{-1} Ni in their above-ground tissues under natural habitat conditions [90,91]. Here, the term "above-ground tissue" should be regarded as plant foliage only and "under natural habitat conditions" indicates that hyperaccumulators must achieve metal hyperaccumulation in a healthy status. Nowadays, except for Ni, more criteria are developed to define hyperaccumulaor for various metals. The basic principle for the evaluation is that metal accumulated in the shoot of hyperaccumulaors should be 100 to 1000-fold higher than that of crops or non-accumulators [92]. The recommended concentration thresholds for different metals in dried foliage matter are shown as follows: 100 mg kg^{-1} for Cd, Se and Tl; 300 mg kg^{-1} for Cu, Cr and Co; 1000 mg kg^{-1} for Pb, As, Ni; 3000 mg kg^{-1} for Zn; and 10000 mg kg^{-1} for Mn [93]. So far, more than 400 species have been identified as metal hyperaccumulators and many of them belong to family *Brassicaceae*, shown in Table 1.

Binomial name	Metal species	Metal accumulation (mg kg^{-1})	Reference
Alyssum markgrafii	Ni	19100	[94]
Alyssum corsicum	Ni	18100	[95]
Alyssum caricum	Ni	12500	
Euphorbia cheiradenia	Pb	1138	[96]
Eleocharis acicularis	As	1470	[97]
Petris vittata	As	8331	[98]
	Cr	20675	
Azolla pinnata	Cd	740	[99]
Thlaspi caerulescens	Cd	263	[100]
Eleocharis acicularis	Zn	11200	[101]
	Cu	20200	
Amaranthus chlorostachys	Cs	2146	[102]
Pennisetum purpureum Schum	Cs	26365	

Table 1: List of some hyperaccumulators.

Another principle for hyperaccumulator exploration is plant's translocation factor (TF) and bioconcentration factor (BCF). The TF and BCF of plant represent the ratio of metal content from shoots to roots and harvested tissues to soils, respectively [103]. It is indicated that plants with both TF and BCF lager than 1 have the preferable potential for phytoextraction. It should be noted that, as the important indicator, the value of TF and BCF can be as high as 50-100, while can also be less than 1 if the metal's concentration in soil was extremely high [104].

Electro-kinetic parameters

Electrode material: Electrode material selection is primarily based on the reactive equilibriums occurred on their surface as well as their inherent thermal, mechanical and corrosive resistance [105]. Many studies have revealed that, the oxide film formed on the electrode surface could significantly block electrode active sites and increase the electric resistance during the EKR process [106]. Thus, electrode pre-coating with material such as platinum, silver, gold and titanium is required in order to increase the roughness and surface active sites of the electrode.

Recently, there is a growing interest in using graphite as electrode material due to their low cost and good accessibility. Since phenomena like oxide film formation and surface precipitation still restrain it lifetime and reactive activity, modifications are always required [107]. Many researchers suggested that forming a thin layer by using metallic oxide on electrode surface (i.e., Ti/SnO_2-Sb_2O_3, carbon/TiO_2, Ti/IrO_2) could be a effective approach as modification [108]. In that way, the modified electrode can be used as both anode and cathode, which is given the name of "Dimensionally Stable Anode" due to its improved resistance to reactive species and pH changes [109].

Electrical field intensity: Electric field intensity is a determining factor in influencing the effectiveness of EK-enhanced phytoextraction. Lemström [60] did the first large-scale experiment to investigate the influence of voltage gradient on plants growth in early 20 th century. In that study, a constant electric field of 10 kV m^{-1} was applied and greener morphological characteristics and dramatic increases in yield were observed from the treated plants. The later studies obtained the similar consequences that electrical field had positive effect on plant's germination and growth rate, while the relative mechanisms were not well explained so far [61]. Except for the benefit, some other scholars also pointed out that damage can be shown on plant's leaves with higher electric field, which indicated that plant's growth is not simply proportional to voltage gradient. Optimization of electrical field intensity for different plant species and the possible damage theories need a further investigation.

It seems that the possible damage theories and optimal electric field intensity for different plant species need further investigation. Cang et al. [62] recently studied the influence of voltage on phytoextraction by using Indian mustard. They found that low electric field was beneficial for Indian mustard's growth since its biomass production was decreased as the increase of voltage. In addition, as the bioavailability of heavy metals was elevated with increasing voltage, the tradeoff for balancing it with voltage negative effects on plant's growth was considered and voltage lower than 2 V/cm was suggested to be preferable in that case [110]. Similar results were also obtained in other studies that under voltage between 0.5-2 V/cm, both plant biomass production and metal accumulation could achieve satisfying performance.

Mode of applied voltage: Previous studies showed increasing bioavailability of soil metals while unfavorable pH polarization under direct current field (DC). The pH polarization, especially alkalization directly restricts the growth of plant as well as the migration of metals [63]. Therefore, alternative current field (AC) which involves a period exchange in polarity direction is studied in order to maintain soil pH in an acceptable range. Aboughalma et al. remediated metal-polluted soils by using potato integrated with electric current AC and DC, respectively [67]. They found that the initial pH value (6.5) was decreased to 3 near the anode regions and increased to 8 near the cathode regions, respectively with application of DC. The target metals had a migration from anode toward cathode while accumulated in the area of pH jumping point. On the contrary, no polarization and sharp transition of soil pH was observed with application of AC. Furthermore, potato had a 72% increase whereas a 27% decline of biomass production under AC and DC, respectively. As a result, potato grew under AC showed higher metal accumulation in both roots and shoots than those in the control and DC test. Bi et al. investigated the phytoextraction under electric field with polarity reversal every 3 h

[83]. Under this voltage mode, they discovered that the soil pH polarization could be totally eliminated and no significant alteration in soil physico-chemical properties was shown. Overall, plant reactions to different electric fields are more or less beneficial for plant biomass production, metal uptake and metal translocation, and the mode of AC shows a better impact on EK-enhanced phytoextraction.

Electrode configuration: Electrode configuration is also an important parameter in influencing the effectiveness of EK-enhanced phytoextraction. Until now, many efforts have been made on this issue: Zhou et al. used a vertical DC which extended the phytoextraction depth and prevented the leaching of Pb Hodlko et al. [111] proposed several electrode arrangements to prevent the leaching of mobilized metals. They suggested the electrode configuration that placed the cathode in the center with anodes surrounded had the greatest potential for metals accumulation [112], In addition, Putra et al. designed a 2D electrode configuration with cathode placed on the top layer of soil and anodes vertically installed in four corners of a rectangular chamber. The results showed enhancement in both metal accumulation and translocation in plants tissues [113]. Based on previous study, electrode configurations are generally divided into unidirectional, and bidirectional and radial types (Figure 3) [114]. They are applied to treat different soil matrix in order to achieve the demanded outcomes. For instance, bidirectional configuration is more effective on metals transportation at high concentration while radial type is more suitable for homogeneous remediation of multi-metals contaminated soil.

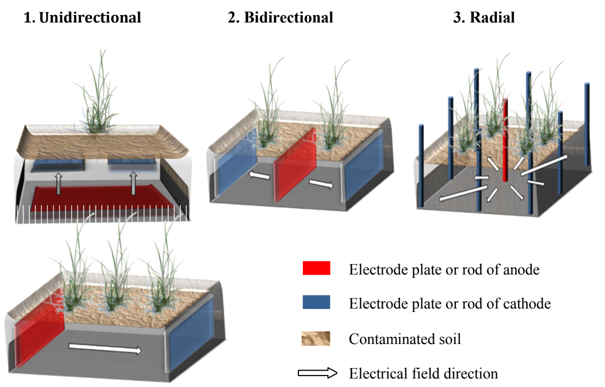

Figure 3: Illustration of 1 Unidirectional, 2 Bidirectional and 3. Radial electrode configurations.

During the electrode configuration designation, many factors such as electrode spacing and cell selection should be carefully considered [111]. Since electrode spacing was found to be inversely proportional to electric field intensity, increasing the electrode distance might cause the reduction of electrical potential and migration of elements, thus extend the remediation period [115]. In addition, selection of experimental setup without constituting electrical conductive material can prevent the short circuit during the EKR [116].

Conclusions

This article clarified and summarized the possible mechanisms and influcing factors of EK-enhanced phytoextraction for remediation of metal-polluted soils. In terms of mechanisms, phytoextraction is a sustainable process for reducing the concentration of metals in soils via various plants. It is a novel, solar-driven efficient, in situ applicable, cost-effective and eco-friendly remediation approach which highly dependent on metal's phytoavailability for its success. EKR involves introducing a direct low-intensity electric field to mobilize target pollutants through the porous medium. The main transport mechanisms for EKR process to mobilize the metal ions include electromigration, electroosmosis and electrophoresis. It is a promising enhancement for increasing metal's phytoavilability as well as plant growth. However, soil pH polarization induced by electrolysis of water during the process still restrict its performance. Thus, strategies like polarity reversal in certain frequency and addition of soil amendments are usually associated with the EK-enhanced phytoextraction in order to keep the soil pH in an acidic range. Regarding to influencing factors, metal's speciation analysis through sequential extraction method is important since only water and acid soluble fractions of metals are available for electromigration or plants' uptake. Based on this procedure, relevant suggestions can be determined for transforming the stable metals to labile fractions. Selection of proper hyperaccumlator which possesses characteristics of high growth rate, high resistance to target metals, remarkable metals accumulation and translocation ability, and good adaptability to environmental and climatic changes is also critical for EK-enhanced phytoextraction. The basic principle for plant's selection include metal's accumulation in the aboveground tissues as well as the TF and BCF of the plant. Moreover, electrical parameters that include electrode material, electrical field intensity, mode of applied voltage and electrode configuration is another important issue. There parameters can highly determine the efficiency of EK-enhanced phytoextraction by influencing the transport-reactive equilibriums and geo-chemical reactions of trace metals in soil matrix. In summary, the mechanisms and influencing factors in the present study are important in the context of the existing literature and for the application and optimization of EK-enhanced phytoextraction as an *in-situ* decontamination. We believe that the contents discussed in this paper can applied as an useful guideline for further investigation of EK-enhanced phytoextraction.

Acknowledgements

This work was supported financially by "the Fundamental Research Funds for the Central Universities" (2017B11014, 2017B20414, 2018B42614), "the China Postdoctoral Science Foundation" (2017M611677) and "Jiangsu Scientific Research Program"(BE2015705, BE2017765). The authors would like to thank all the beneficiaries of the project for the support and assistance provided.

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