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The definitive publisher version is available online at <https://doi.org/10.1016/j.jhazmat.2020.123891>

1 **Enhanced copper removal from contaminated kaolinite soil by**
2 **electrokinetic process using compost reactive filter media**

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21 **Abstract**

22 Electrokinetic (EK) remediation is a promising technology for soil decontamination, although
23 basic pH in the soil close to cathode has constrained EK effectiveness due to heavy metal
24 precipitation. This study aimed to enhance copper removal from kaolinite soil by integrating
25 EK with compost (C) as recyclable reactive filter media (RFM) for the first time. Compost
26 placed near the cathode served as an adsorbent to bind copper ions while buffering the
27 advancement of the alkaline front in soil. The total copper removal rate increased from 1.03%
28 in EK to 45.65% in EK-100%C under an electric potential of 10 volts. Further experiments
29 conducted by using biochar (BC) and compost/biochar (C+BC) mixture RFM at different ratios
30 showed total Cu removal efficiency decreasing as EK-100%C > EK-(10%BC+90%C) > EK-
31 (20%BC+80%C) > EK-(30%BC+70%C) > EK. The application of a constant electric current
32 of 20.00 mA further enhanced copper removal to 84.09% in EK-100%C although did not show
33 significant enhancement in EK-(BC+C). The compost RFM was regenerated by acid extraction
34 and then reused twice, achieving a total removal of 74%. The findings demonstrated compost
35 as a promising and reusable RFM for the efficient removal of copper in contaminated soil.

36

37 *Keywords:* Electrokinetic; Soil decontamination; Copper; Reactive filter media, Inorganic
38 pollutants

39 **1. Introduction**

40 Soil contamination by heavy metals is a global environmental issue and may cause serious
41 environmental risk and potential harm to human and living beings [1,2]. For example, soil
42 samples from a former wood impregnation site north of Copenhagen, Denmark were found to
43 contain 1662 mg kg⁻¹ of copper among a suite of heavy metals [3]. Similarly, as high as 800
44 mg kg⁻¹ of copper was detected in soils from vineyards in France [4]. Therefore, remediation
45 technologies that ensure the removal of heavy metals with the least damage to the soil
46 environment are urgently sought. Electrokinetic (EK) treatment is an emerging technique for
47 heavy metals removal from soils, especially those with low permeability which are usually
48 difficult to be treated [5–8], by imposing a small electric field between electrodes installed in
49 the contaminated soil [7,9,10]. Heavy metals are typically extracted from soil mainly by
50 electromigration mechanism during the EK remediation treatment. The removal of heavy metal
51 contaminants from the soil in EK is significantly pH-dependent, as an alkaline front is
52 developed which is responsible for the precipitation of heavy metal ions close to the cathode.
53 This alkaline condition would lead to the sorption of the heavy metals on soil particle surfaces
54 and/or their precipitation in the soil pores, which may make the heavy metals immobile
55 temporarily and difficult to remove [11]. The application of EK remediation can only remove
56 mobile heavy metals from soil sections near the anode so that heavy metals precipitate in the
57 soil close to the cathode zone, where base and acid fronts meet. Therefore, there is an increasing
58 demand to develop enhanced EK techniques, which can mitigate the advancement of the
59 alkaline front by providing the desired soil pH for heavy metals removal [1].

60 A low pH condition would enhance the removal of heavy metals from the contaminated
61 soils, by improving desorption of the majority of heavy metals from the surface of soil [12]. In
62 EK laboratory experiments, the catholyte compartment was treated by applying an
63 enhancement agent such as surfactants, chelating agents [13,14] or acids/bases [5,12,15], to
64 increase the contaminant mobility or adjust the pH. Bahemmat et al. [16] investigated the use

65 of fulvic and humic acids as chelating agents as well as the effects of catholyte conditioning to
66 improve EK remediation efficiency in highly heavy metals contaminated soil. They indicated
67 that both humic substances and catholyte conditioning with 0.1 N HNO₃ enhanced the
68 remediation efficiency of heavy metals by two to three times after 20 days, compared to the
69 conventional EK treatment. Yuan et al. [1] proposed an enhanced EK remediation using citric
70 acid + CaCl₂ for the removal of heavy metals from a contaminated kaolinite soil. The study
71 reported an improved remediation process compared to other agents such as HCl in terms of
72 metal removal, power consumption and environmental risk. In studying two different
73 conditioning electrolytes (polyaspartic acid, citric acid) for EK remediation of industrial
74 chromium waste, Fu et al. [17] showed that citric acid was merely effective for total Cr removal.
75 Moreover, energy expenditure was increased in EK remediation by adding the enhancement
76 agents because of water electrolysis, heat loss and ions transport in comparison with deionized
77 water as the electrolytes [17]. Iannelli et al. [5] investigated the effect of conditioning agents
78 such as citric acid, oxalic acid, ascorbic acid, HCl, HNO₃, H₂SO₄, and EDTA for the extraction
79 of heavy metals (Cd, Cr, Cu, Ni, Pb, and Zn) from marine sediments. They reported that
80 although sulphuric acid enhanced the removal efficiency, it produced significant precipitation
81 of sulphates in the solid material and catholyte. Also, HCl acid was found to be the most
82 effective for the metals; however, significant chlorine gas was produced at the anode which
83 required further gas treatment [5].

84 The review demonstrated that although strong acids could overcome the major drawback
85 of EK process by preventing the advancement of the alkaline front in soil, they would increase
86 the power consumption and treatment duration [12]. Moreover, some soil properties such as pH
87 and electrical conductivity may be altered by the addition of the enhancement agent [16].
88 Additionally, the electrolyte should ideally be recovered after EK treatment, which will increase
89 the remediation cost [11,18]. Some enhancement agents may not be practical for the removal
90 of heavy metals [19], and their application in full-scale EK remediation may often be

91 unsuccessful [5,19]. Besides the enhancement agents, there are hybrid techniques to extract the
92 heavy metals, such as EK-phytoremediation [20,21], EK-bioremediation [22], and EK
93 remediation coupled with a permeable reactive barrier (PRB) [23,24]. However, among these
94 combined techniques, PRB can facilitate the removal of heavy metals with a short operation
95 period and prevents the contamination of the catholyte solution [23,25], which make the
96 technique cost-effective. PRB serves as an adsorbent during the electromigration and
97 electroosmosis process to trap the metals ions transported from the contaminated soil.

98 To overcome the limitations of existing approaches using chemical agents in EK process,
99 a permeable reactive barrier (PRB) made of activated carbon (AC), activated bamboo charcoal,
100 and zero-valent iron [23,24,26,27] have been used, which have shown better performance
101 compared with the conventional EK process. However, their application would be restricted
102 due to their cost, availability, and especially life cycle. Besides, PRBs may not be able to
103 prevent the advancement of the alkaline front, and hence their application was always combined
104 with chemicals. As a result, EK treatment will not be a cost-effective process [26,28].

105 This study, therefore, aims to use environmental-friendly reactive filter media (RFM) to
106 improve Cu removal from contaminated kaolinite soil. Copper was used as the target
107 contaminant because of its wide occurrence in the soil environment. Specifically, the first
108 objective is to apply compost as a novel and recyclable RFM near the cathode to buffer the
109 alkaline front and to capture copper ions. Compost is widely applied to treat the contaminated
110 soil and groundwater due to its low-cost and adsorption capacity of humic substances [29–31];
111 however, no application of compost in the EK process has been reported yet. The application
112 of compost RFM in EK process presents several advantages. First compost is rich in organic
113 matter and nutrients and often used to improve soil fertility and quality [28]. It is of low cost
114 and environmentally friendly. The humic substances and iron oxides in compost can form stable
115 complexes with heavy metal ions, helping in their metal uptake from contaminated soil [32].
116 Additionally, the potential regeneration and reuse of compost RFM were evaluated for copper

117 removal. The second objective is to use a mixture of biochar and compost at different ratios as
118 a novel RFM in the EK-RFM system. Similar to compost, biochar is widely used as a soil
119 conditioner and has shown a high capacity for heavy metals adsorption from the contaminated
120 soil [33,34]. The performance of biochar filter media in the EK process has been investigated
121 in a previous study while improving the extraction of copper from the contaminated kaolinite
122 soil [25]. The potential drawbacks of compost and biochar are their highly complex composition
123 and relatively small particle size, hence both need to be packed in RFM to prevent their loss
124 into the soil. At the same time, their efficiency in Cu removal has to be determined. The findings
125 would provide further insights into the best RFM for effective removal of heavy metals from
126 soil using EK remediation technology.

127

128 **2. Materials and methods**

129 *2.1. Materials, soil preparation and analysis*

130 Copper sulphate (CuSO_4) with a purity > 99% was purchased from Sigma-Aldrich, Australia.
131 To prepare a stock solution of $1000 \text{ mg L}^{-1} \text{ Cu}^{2+}$, a given amount of copper sulphate (2.52 g)
132 was dissolved in 1000 mL of distilled water. The chemical and physical characteristics of the
133 commercial kaolinite soil, used in this study, were reported [25]. Briefly, copper-contaminated
134 kaolinite soil was prepared by thoroughly mixing 1000 g of kaolinite with 1000 mL of an
135 aqueous solution containing 1000 mg L^{-1} copper. The copper-contaminated kaolinite soil was
136 loaded into the EK cell after 72 h [35]. All the tests were performed with an initial moisture
137 content of 100%. Compost (Searles 10 L, Organic Compost), purchased from Bunnings
138 Warehouse Ltd (Australia), was used as supplied. The detailed properties of compost are shown
139 in Table 1. Compost (70 g) was used to fully pack the EK cell RFM between soil and cathode
140 compartment. Copper concentration in both soil and RFM was measured by a portable X-ray
141 Fluorescence (XRF) instrument, as used by Ghobadi et al. [25]. The advantages of portable
142 XRF compared to the traditional laboratory techniques such as ICP-MS are the limited

143 preparation required for solid samples, non-destructive analysis, decreased production of
144 hazardous waste, low start-up and running costs, and increased total speed and throughput [36–
145 38]. Samples of the contaminated soil and RFM were first dried, pulverized using a mortar to
146 be in the form of loose powders, and then filled in the XRF sample cups for Cu concentration
147 analysis [39,40]. The procedure for the elemental analysis of Cu was conducted by Vanta
148 software inserted in the XRF instrument, which is regularly calibrated. The pH and electrical
149 conductivity of kaolinite soil before and after the EK treatments were determined using a
150 multimeter (model Hach HQ40d) by providing slurries with dry soil to water ratio of 1:5 (w/v)
151 [35].

152 In the compost regeneration experiments, the compost was removed from the EK cell and
153 mixed with a known amount of concentrated HCl acid solution in a 250-mL Erlenmeyer flask
154 [41]. The flask was then placed in an orbital shaker incubator (Thermoline Scientific) to
155 properly shake at 250 rpm and room temperature for 24 h. After that, the sample was centrifuged
156 to separate compost from the acid solution, and the compost was washed by deionized water to
157 adjust the pH to its original condition. Then, the sample of the compost RFM was dried and
158 grounded [40] for the copper concentration measurement by using the handheld XRF analyzer
159 before reuse.

160 In addition, a novel recyclable RFM was designed for soil remediation by mixing the
161 compost with biochar at different ratios to increase the RFM adsorption capacity. The previous
162 experimental results have proved biochar as a potential RFM to enhance copper extraction from
163 contaminated kaolinite soil in the EK system [25]. However, the performance of EK-BC
164 required further improvement. Therefore, commercial biochar (purchased from Karr Group
165 Co.) used in our previous study [25], was mixed with the compost at different ratios while
166 maintaining the same total amount of RFM at 70 g in all experiments. The RFM blends were
167 applied in a series of EK-RFM operations to examine the feasibility of copper extraction from
168 kaolinite soil. A summary of the physicochemical characteristics of RFM is shown in **Table 1**.

169 The morphological and chemical composition of compost RFM were determined by using
 170 Energy Dispersive X-Ray Spectroscopy (EDX), which was a chemical microanalysis
 171 technique, used in conjunction with scanning electron microscopy (SEM) (model Zeiss Evo-
 172 SEM). A Nano-ZS Zeta-seizer (Malvern, Model: ZEN3600) was used to measure the zeta
 173 potential of the compost before and after treatment. In addition, Fourier Transform Infrared
 174 Spectroscopy (FTIR) (Miracle-10: Shimadzu) analyses were performed to characterize surface
 175 functional groups for both treated and untreated RFMs.

176

177 **Table 1.** Physicochemical characteristics of RFMs.

Properties	Compost	Biochar
Particle size distribution (%)		
d > 2 mm	21.50	2.59
1 mm < d < 2 mm	21.70	14.51
d < 1 mm	56.80	82.89
Permeability (m s ⁻¹)	0.72×10 ⁻³	0.17×10 ⁻³
Organic matter (%)	43.09	98.36
pH	5.13±0.20	8.95±0.05
Electrical conductivity (mS cm ⁻¹)	1.11	0.11
Metals ion concentration (mg kg ⁻¹)		
Cu	135	9
Cd	12	21
Zn	122	35
Pb	8	3
Elemental composition (%)		
O	50.45	11.60
C	35.02	87.23

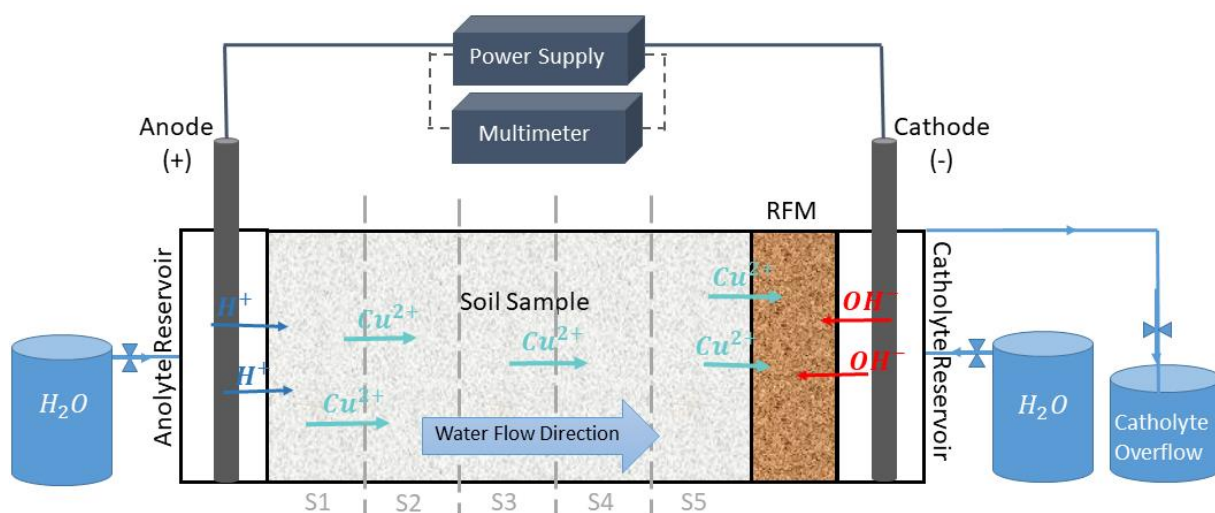
N	7.78	N/A
Si	4.76	0.37
Al	1.50	N/A
Other	0.49	0.80

178

179 2.2. EK apparatus and experimental design

180 **Fig. 1** illustrated a schematic diagram of the EK set-up, consisting of a reactor made of
 181 plexiglass material with dimensions of 230 mm × 80 mm × 110 mm. The reactor was composed
 182 of six parts: soil compartment, RFM compartment, electrode compartments, electrolyte
 183 reservoirs, electrolyte overflow reservoir, and power supply. A pair of graphite rod electrodes
 184 (150 mm × 10 mm) were placed in each electrode chamber to apply the electric field by a power
 185 supply. A current meter was used to monitor the electric current. RFM was packed in the EK
 186 reactor, between the soil and the cathode compartments. A filter paper (pore size 5-13 μm, LLG
 187 Labware) was used to separate the RFM from the soil. Two filter papers were also attached to
 188 a pierced plexiglass plate, one inserted between the soil and the anode chamber and the other
 189 one placed between RFM and the cathode compartment, to prevent soil and RFM dispersion or
 190 leakage to the electrolyte cells. Deionized water as electrolyte solution was fed into both anode
 191 and cathode compartments.

192



193

194 **Fig. 1.** Experimental setup for EK-RFM remediation test.

195

196 Ten EK experiments were carried out at room temperature, without pH control for a
197 treatment time of seven days to evaluate copper extraction from the contaminated kaolinite soil.
198 Experimental conditions are summarized in **Table 2**. A fixed voltage of 10 V was imposed on
199 the saturated soil in Exp-01 to Exp-06, while Exp-07 to Exp-10 were conducted under a fixed
200 electric current (20.00 mA).

201 Exp-01 was the conventional EK treatment and performed as a reference (unenhanced)
202 experiment to investigate copper extraction by using deionized water as the electrolyte. Exp-02
203 was implemented to investigate the effect of compost RFM on the EK removal of copper from
204 the kaolinite soil. To design a novel recyclable RFM, a series of EK-RFM experiments (Exp-
205 03 to Exp-05) were performed by mixing compost with biochar in different ratios aiming to
206 increase the RFM adsorption capacity for copper removal. An additional EK-RFM test (Exp-
207 06) was also run by loading biochar (100%BC) in the RFM compartment for comparison of the
208 performance of RFMs in the EK-RFM process. The weight of RFM for all EK-RFM tests was
209 70 g.

210 Exp-07 and Exp-08 were performed, applying a fixed electrical current (20.00 mA) for the
211 treatment of EK-C and EK-(BC+C). To examine the regeneration of compost RFM and the
212 performance of the recycled compost RFM in the EK process, after Exp-07, compost RFM was
213 recovered by acid solvent extraction technique and reused in the EK-C treatments (Exp-09,
214 Exp-10) in two cycles.

215 After seven days, the DC source was disconnected, and the set-up was disassembled. At
216 the end of each test, the soil sample was cut into five equal sections, S1 to S5, from the anode
217 to cathode. Each slice was oven-dried and analyzed for pH, electrical conductivity and copper
218 content. All data were inputted into OriginPro (v 9.6), and an ANOVA test was conducted with
219 a significance level of 0.05 to differentiate **between** the EK treatments.

221 **Table 2.** Experimental condition of different EK experiments.

Exp. no.	Experiment type	RFM	Constant voltage (V)	Constant current (mA)	Cu (mg kg ⁻¹)	Operation time (d)
Exp-01	EK	N/A	10		1000	7
Exp-02	EK-100%C	Compost	10		1000	7
Exp-03	EK- (10%BC+90%C)	Biochar+Compost	10		1000	7
Exp-04	EK- (20%BC+80%C)	Biochar+Compost	10		1000	7
Exp-05	EK- (30%BC+70%C)	Biochar+Compost	10		1000	7
Exp-06	EK-100%BC	Biochar	10		1000	7
Exp-07	EK-100%C	Compost		20.00	1000	7
Exp-08	EK- (10%BC+90%C)	Biochar+Compost		20.00	1000	7
Exp-09	EK-RC	Recycled Compost		20.00	1000	7
Exp-10	EK-RC	Recycled Compost		20.00	1000	7

222

223 **3. Results and discussion**224 *3.1. Performance of EK-C in copper removal*225 *3.1.1. Profiles of electric current, soil pH and electrical conductivity*

226 **Fig. 2a** presents the change of electric current with time, **under a constant applied voltage of**
227 **10-V**, for the EK-C as well as the conventional EK experiment. Generally, in the EK

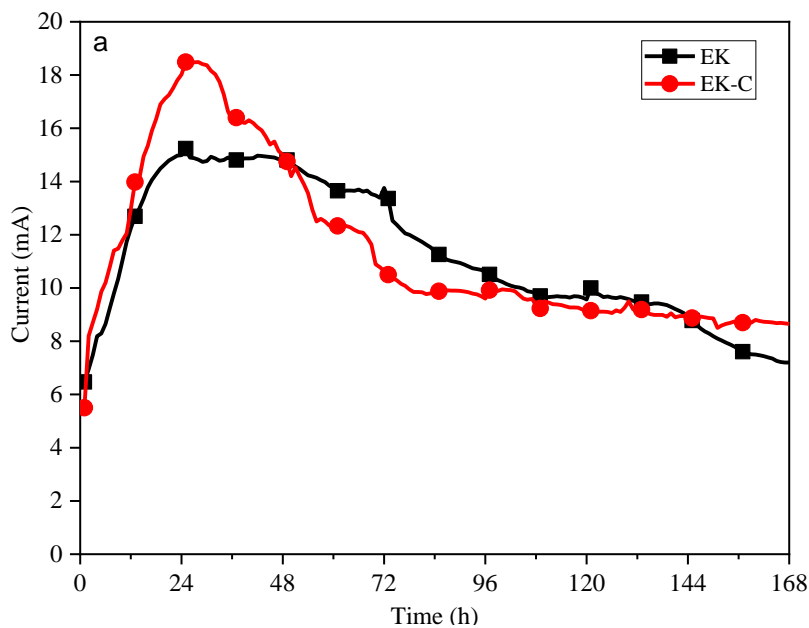
228 remediation of the heavy metal-contaminated soil, the electric current increased firstly, then
229 reached its highest value after a certain time due to the accelerated generation of the acid front
230 at the anode, leading to the solubilization of copper ions and their migration through the soil in
231 the direction of the cathode [25,42]. Afterwards, the electric current slowly decreased and
232 stabilized at a low constant value, which could be attributed to the reduction of mobile ions in
233 the soil [17,25,43]. The alkaline pH would explain the continuous decrease of the electric
234 current at the cathode side. The development of the alkaline front into the soil resulted in the
235 precipitation of metals and fewer ions available for the transportation of the electric charge [44].
236 Besides, the decrease in the content of mobile ions in the soil refers to the movement and
237 extraction of free metals [17]. The pattern of current change in time in the EK-C experiment is
238 very similar to that in the previous studies [25] (**Fig. 2a**). **The average electric current for the**
239 **EK-C test (11.42 mA) was found to be slightly higher than that in the EK treatment (11.30 mA).**
240 The higher electric current would result in a higher metal contaminant removal (**Fig. 3**), and it
241 was due to the presence of compost filter media in the EK system, which buffered the advance
242 of hydroxide ions generated at the cathode reaction, promoting the transport of the acid front in
243 the soil. In the EK-C experiment, electric current was at a low value of 5.50 mA in the
244 beginning, and then reached a maximum value of 18.49 mA within a day. After that, it
245 dramatically dropped to 10.64 mA over 72 h and remained almost stable at 8.65 mA by the end
246 of the experiment. The sharp fall may be associated with the early precipitation of copper ions
247 in the soil. The highest value of electric current in the EK experiment without compost RFM
248 was 14.97 mA. As shown in **Fig. 2a**, the EK-C test also showed a higher electric current at the
249 end of the experiment in comparison with that of the EK without RFM. This could be explained
250 by the continuous adsorption of OH⁻ produced at the cathode reaction by the compost,
251 promoting the advancement of the acid front in the soil and hence maintaining the electric
252 current at a higher level.

253 The available form of heavy metals in the soil is significantly pH-dependent. Hence, a low
254 pH environment can enhance the heavy metals dissolution and the degree of their removal from
255 the contaminated soil [43,45]. When inducing the electric field during the EK process, the soil
256 pH would change due to the chemical reactions that occurred in the EK system [46,47].
257 Specifically, the application of the electrical field causes the oxidation of water at the surface
258 of the anode, which causes a reduction in pH due to the production of H⁺ ions. Hence, an acid
259 front is electrogenerated at the anode region favoring the dissolution of the heavy metals
260 migrated towards the cathode [44,47,48]. At the same time, an increase in the soil pH near the
261 cathode side is derived from the furtherance of OH⁻ created by the reduction of water at the
262 cathode surface [47]. These changes in pH would lead to dissolution or precipitation of heavy
263 metal contaminants across the soil. **Fig. 2b** presents the soil pH profile in different soil sections
264 from the anode to cathode (S1-S5) at the end of EK experiments. Compared to the pure EK
265 treatment, the EK-C remarkably presented a lower soil pH, due the compost RFM adsorbed the
266 hydroxide ions produced at the cathode migrating towards the anode. Lower pH in the soil near
267 the cathode region would cause less Cu precipitation in S5, and consequently, copper removal
268 would be achieved in a reasonable degree. In the EK-C test, the soil was acidic through S1 to
269 S4, ranging between pH 2.92 and pH 3.35, which favored copper removal in those sections.
270 Soil pH in S5 near the cathode region was only 5.70, slightly above the initial soil pH, while it
271 was about 9.29 in the EK test without RFM. Compost RFM could successfully lower the soil
272 pH in the soil sections close to the cathode side, from pH 4.36 to pH 3.35 in S4 and from pH
273 9.29 to pH 5.70 in S5, which is usually high, at alkali condition, when there is no pH control or
274 RFM in the EK experiment. The high pH area near the cathode in the conventional EK caused
275 massive precipitation of Cu(OH)₂ [6,49], which blocked the pore fluid and made copper
276 removal difficult during the treatment.

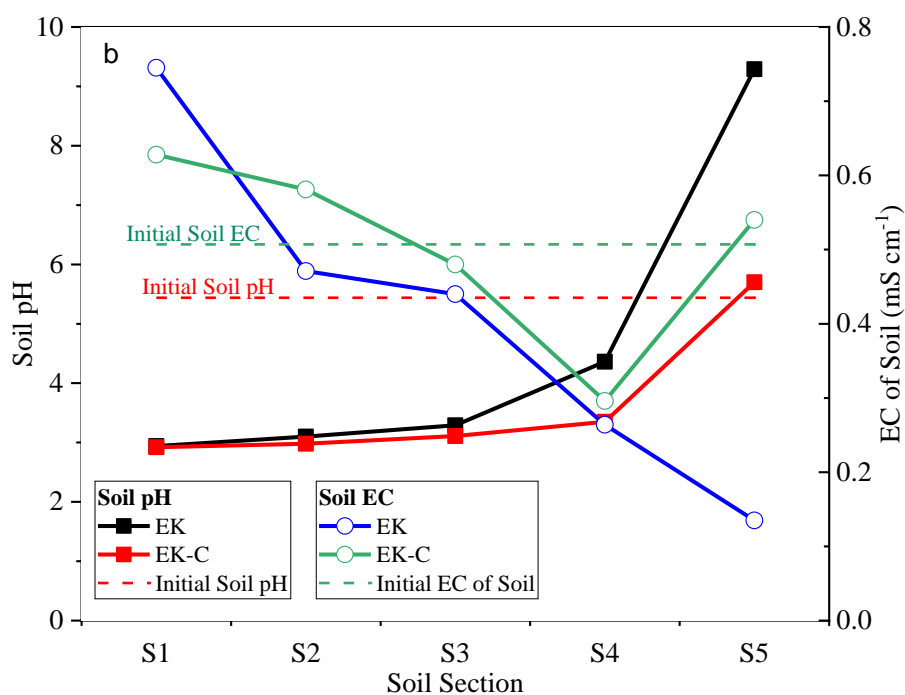
277 **Fig. 2b** also shows the electrical conductivity of soil sections (S1-S5) after the EK
278 treatments. The soil EC values in the EK-C experiment were higher than that in the EK test

279 without compost RFM. This tendency was following the change of electrical current and soil
280 pH, as explained above. The electrical conductivity of the soil in the conventional EK treatment
281 represented a decreasing trend from anode towards cathode; however, the behavior was
282 inconsistent in the EK-C as the EC increased in S5 near the cathode. In the conventional EK
283 test, the hydroxide ions moved from the cathode area into the soil and reacted with copper ions
284 in soil pore fluid to form $\text{Cu}(\text{OH})_2$ precipitation [6,49]. As a result, the EC of soil decreased
285 near the cathode area. In contrast, the presence of compost RFM in the EK-C treatment could
286 successfully reduce the formation of copper hydroxide in S5 near the cathode, which resulted
287 in higher soil EC in S5. In general, compared with other types of RFM such as activated carbon
288 and biochar, which were investigated in our previous work [25], compost had the greatest
289 capacity in the EK-RFM process in buffering soil pH as well as presenting higher soil EC (**Fig.**
290 **2b**).

291



292



293
 294 **Fig. 2.** (a) Variation of electric current during seven days of EK operations under a constant
 295 voltage; (b) profiles of pH and electric conductivity in soil sections (from anode to cathode)
 296 after the experiments.

297
 298 **3.1.2. Copper extraction from soil**

299 Metal ions during EK treatment are mainly transported through electroosmosis and
 300 electromigration processes [50]. Generally, the ionic species move towards the opposite-
 301 charged electrode [51] as anticipated, hence copper cations electromigrated from anode to
 302 cathode (**Fig. 3a**). The advancement of the acid front at anode resulted in the dissolution of Cu
 303 into the pore solution as Cu^{2+} , which then transported towards the cathode. However, the
 304 electromigration of Cu^{2+} was retarded by the alkaline pH front at the cathode, which caused Cu
 305 accumulation in the soil section near the cathode (S5).

306 In general, the results of Cu^{2+} concentration in the soil were consistent with those of electric
 307 current, soil pH, and soil electric conductivity. After EK-C remediation, the final copper content
 308 in the soil was lower than the initial ones. According to **Fig. 3**, EK-C operation was found to
 309 be more successful in removing copper from the kaolinite soil compared to the conventional

310 EK test. In the EK-C experiment, the copper concentration declined enormously from the initial
 311 content (1000 mg kg⁻¹) to 105 mg kg⁻¹ in S1, and then gradually raised to about 496 mg kg⁻¹ in
 312 S4. Copper precipitation has been significantly decreased in S5 close to the cathode
 313 compartment (1441 mg kg⁻¹) due to lower soil pH in EK-C (**Fig. 2b**), compared to that of the
 314 EK without RFM, which are significantly different among each other ($P < 0.05$). It can be
 315 observed from **Fig. 3a** that more than half of copper usually accumulated in S5 during the
 316 conventional EK test was successfully trapped by compost RFM in the EK-C treatment.
 317 Moreover, copper concentration in other sections has been considerably reduced, particularly
 318 in S4. Which can be ascribed to the decreasing soil pH in the EK-C operation (**Fig. 2b**).

319 The efficiency of the EK treatments was also evaluated by calculating copper removal
 320 efficiency in each soil section using Eq. (1) and presented in **Fig. 3b**:

$$321 \quad \text{Removal efficiency}_i = ((m_{i,initial} - m_{i,final})/m_{i,initial}) * 100\% \quad (1)$$

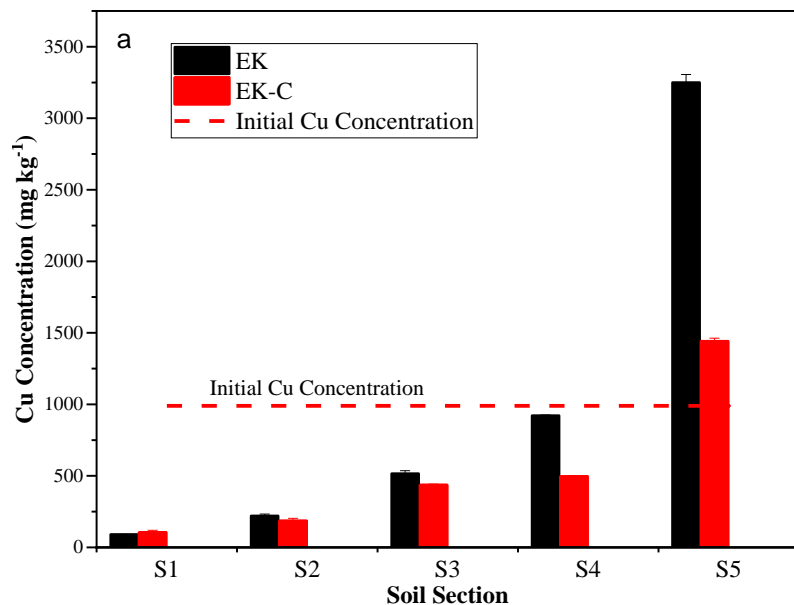
322 where $m_{i,initial}$ is the initial copper concentration (mg kg⁻¹) in section I, and $m_{i,final}$ is the
 323 residual copper (mg kg⁻¹) in section i after the experiment. The negative value indicates that
 324 copper is accumulated in that section. EK-C and conventional EK operations represented almost
 325 similar performance in Cu removal in S3-S1, ranging from 54.10% to 91.02% (**Fig. 3b**).
 326 However, the percentage of Cu removal in S4 (47.79%) and S5 (-51.68%) for the EK-C was
 327 significantly higher than that in the EK without RFM, at 8.69% and -222.20%, respectively,
 328 which are statistically significantly different ($P < 0.05$).

329 Total copper removal from the soil sample is an important parameter, indicating the EK
 330 treatments performance, which is determined from Eq. (2) [52]:

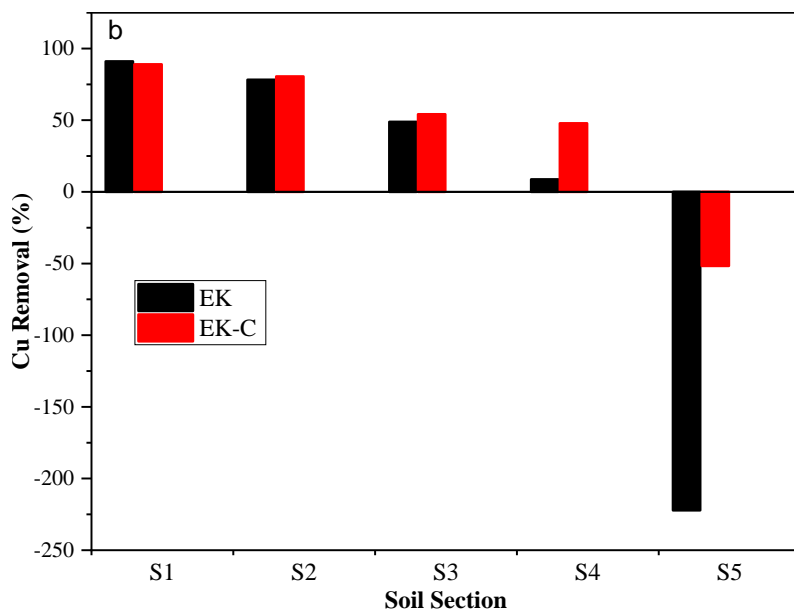
$$331 \quad \text{Total Cu removal} = ((m_{initial} - m_{final})/m_{initial}) * 100\% \quad (2)$$

332 where $m_{initial}$ is the initial copper mass (mg) in the whole soil sample, and m_{final} the final
 333 residual copper mass (mg) in the soil after treatment. The total copper removal rate for the EK-
 334 C test reached the highest of 45.65% (**Table 3**). This rate was reported at the lower values for
 335 the EK coupled with other types of RFM such as AC and BC at the same experimental

336 conditions, 10% and 27%, respectively [25]. In contrast, total copper removal was 1.03% in the
 337 conventional EK test due to the massive copper accumulation in S5 (close to the cathode zone)
 338 in the latter test. The comparison between the recent results and those from previous work [25]
 339 confirms that compost RFM has the highest adsorption capacity for copper without adding
 340 chemicals, preventing additional pollution by the electrolytes and their further treatment.



341



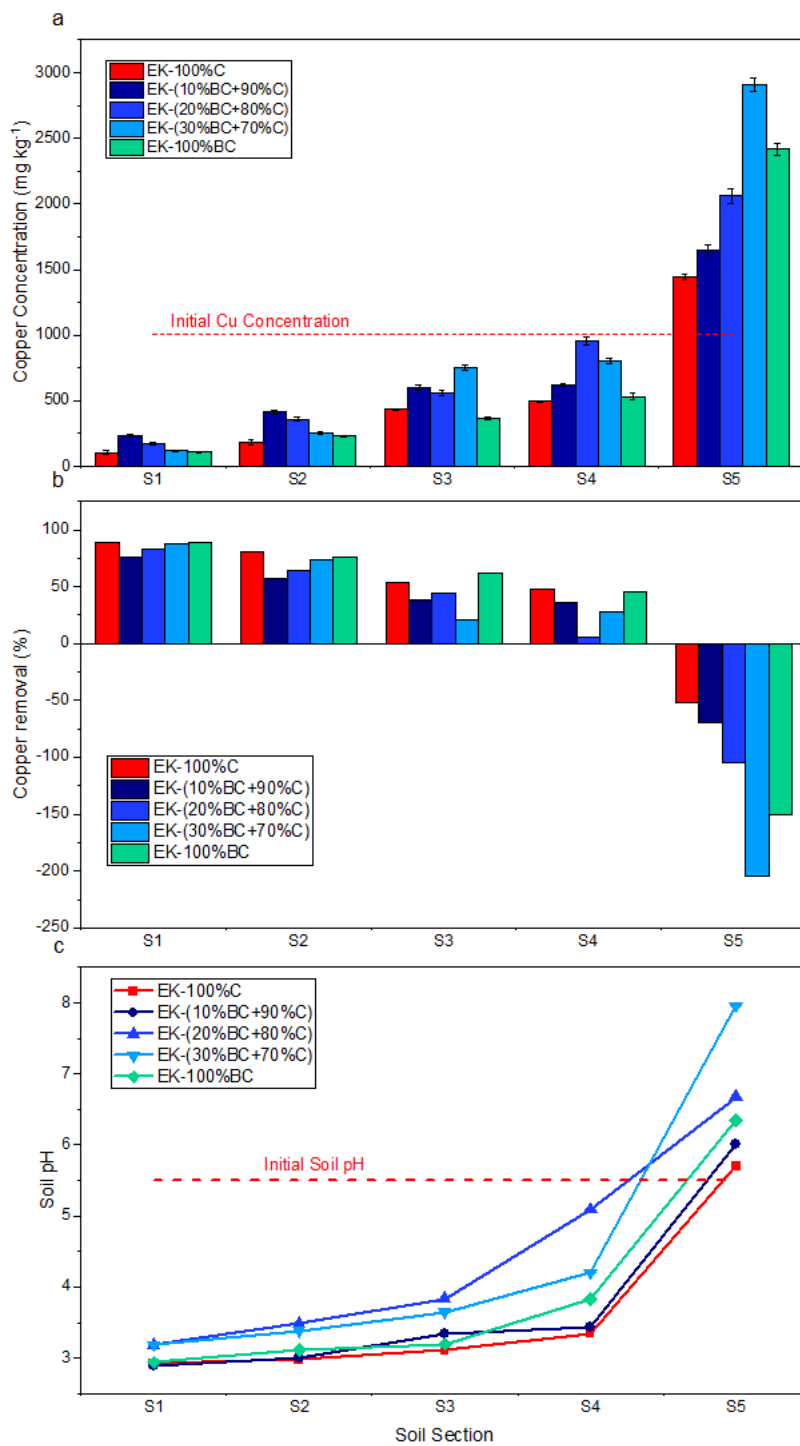
342

343 **Fig. 3.** (a) Residual copper and (b) efficiency of copper removal in soil sections S1-S5 after EK
 344 and EK-C experiments.

345

346 **3.2. Performance of EK-(BC+C) in copper removal**

347 **Fig. 4** presents copper concentration, copper removal efficiency and soil pH in the soil sections
348 from the anode to the cathode for the EK-(BC+C) experiments at the different biochar-compost
349 ratios. For comparison, the results of the EK-100%C test and EK-100%BC treatment were also
350 displayed in **Fig. 4**. In general, the removal efficiency of the EK-(BC+C) increased with the
351 increasing percentage of compost in the RFM mixture, particularly in the soil portions near the
352 cathode area (**Fig. 4b**) while decreasing the copper concentration (**Fig. 4a**). Adding biochar to
353 compost RFM increased the soil pH during the EK-RFM treatment, especially in the areas close
354 to the cathode zone (S5) (**Fig. 4c**). It would retard the impact of humic substances of compost
355 in lowering soil pH and binding metal ions from the contaminated soil. Total copper removal
356 of the EK-(BC+C) experiments was following a declining order: EK-(10%BC+90%C) > EK-
357 (20%BC+80%C) > EK-(30%BC+70%C), at 31.01%, 19.94% and 7.67%, respectively (**Table**
358 **3**). In the case of EK-100%BC, 26.83% of copper was removed during the treatment. All RFMs
359 reduced the availability of copper in the soil during the EK-RFM process, and 100%C had the
360 greatest capacity for that. The reason could be that compost was capable of buffering the
361 alkaline soil pH close to the cathode region, which enhances the efficiency of RFM in copper
362 removal.



363

364 **Fig. 4.** (a) Copper concentration, (b) removal efficiency of copper, and (c) pH profile across the
 365 soil sections at the end of EK treatment.

366

367 **3.3. Characterization of compost RFM**

368 To validate the results presented above, compost RFM samples were taken after the EK
 369 treatment to be characterized to understand their composition better. It was performed by using

370 energy dispersive X-ray spectroscopy coupled with scanning electron microscopy (EDS-SEM)
371 (**Fig. 5**) to confirm the capacity of compost RFM in trapping Cu ions from the contaminated
372 soil in the EK process. **Fig. 5b** shows a typical selected area for EDS mapping measurement,
373 and the EDS analysis indicates that the sample contains Cu element, which is homogeneously
374 distributed in the compost RFM sample (**Fig. 5c**). Besides, the survey spectrum again confirms
375 the presence of Cu in compost samples after EK treatment (**Fig. 5a**).

376 The EK phenomena significantly affected the zeta potential of the compost RFM. Zeta
377 potential of compost increased from -6.39 mV to -0.77 mV after the EK treatment (**Fig. 6a**).
378 This increase is probably due to the interaction between compost particles and free Cu ions.
379 The adsorption of the positively charged heavy metal cations existing in the soil decreases the
380 negative charge of compost particles; therefore, zeta potential becomes less negative.

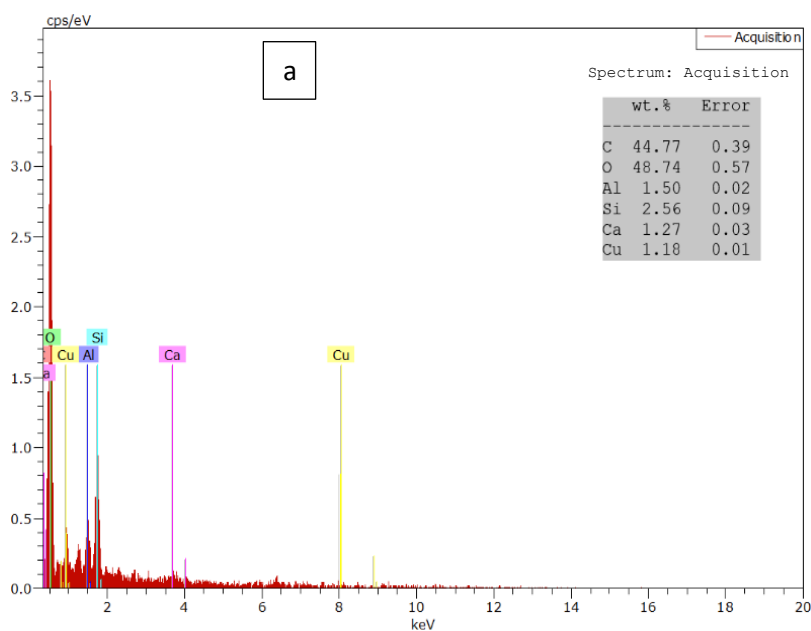
381 FTIR spectra were applied as an analytical technique for providing valuable information
382 on the functional characteristics of compost RFM before and after EK operation (**Fig. 6b**).
383 Although the characteristic peaks of the main organic components are present in both spectra,
384 significant differences can be observed to some extent. There were some major changes, such
385 as the appearance or disappearance of peaks were observed in the EK treated compost sample
386 when compared to the reference (compost before the EK treatment). Also, in **Fig. 6b**, FTIR
387 spectra of compost before and after the EK operations were compared with those in biochar
388 RFM. The FTIR spectrum of both RFMs displayed the band centered at approximately 1630
389 cm^{-1} , which can be ascribed to the C=O stretching, mainly in the carboxyl groups [53]. A strong
390 band at 1030 cm^{-1} which was observed only in the spectrum of compost RFM could be related
391 to the C-O-C bond in polysaccharides; however, the peak increased after EK treatment, which
392 can be attributed to copper absorption. O-H stretch occurred as a very broad peak at 3000 to
393 3500 cm^{-1} in the FTIR spectrum of the compost. The intensity of this band increased in the
394 spectrum of the treated compost, which can contribute to the capacity of the compost in
395 buffering soil pH during the EK process. Comparison of chemical composition (**Table 1**) and

396 the FTIR spectra of compost and biochar RFMs after EK treatment indicated that the oxygen-
397 containing functional group could be involved in the copper ion adsorption by the compost
398 RFM in the EK-C treatment.

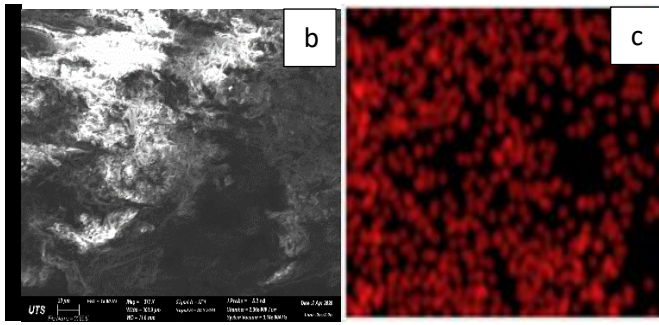
399 In general, adsorption of Cu in compost, which contains a wide variety of organic
400 compounds, could involve several possible mechanisms such as (1) Cu ions exchange with
401 Ca^{2+} , Mg^{2+} and other cations in the compost, ascribing to co-precipitation inner-sphere
402 complexation with complexed humic matter and mineral oxides of compost; (2) the surface
403 complexation of Cu ions with different functional groups, and inner-sphere complexation with
404 the free hydroxyl of mineral oxides and other surface precipitation; and (3) the physical
405 adsorption and surface precipitation. According to the experimental results, it was found that
406 compost RFM had the greatest efficiency of adsorbing Cu, compared to the mixture of
407 biochar/compost and biochar itself. This presumably is attributed to higher O-content and
408 surface functional groups in the compost, compared to biochar, which was related to sorption
409 of Cu ions.

410

411



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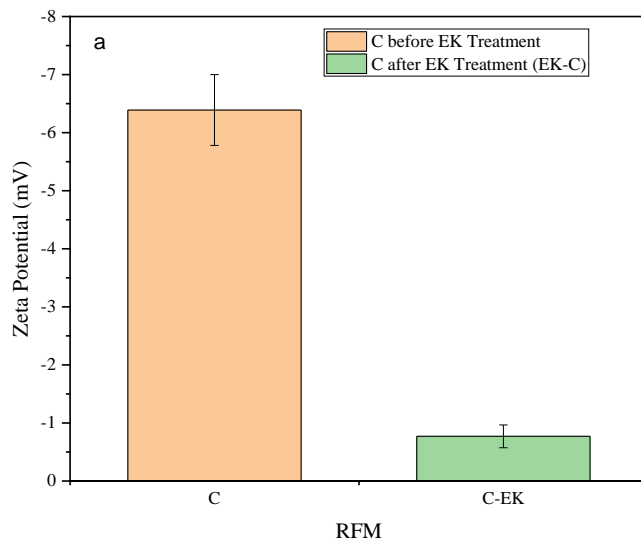


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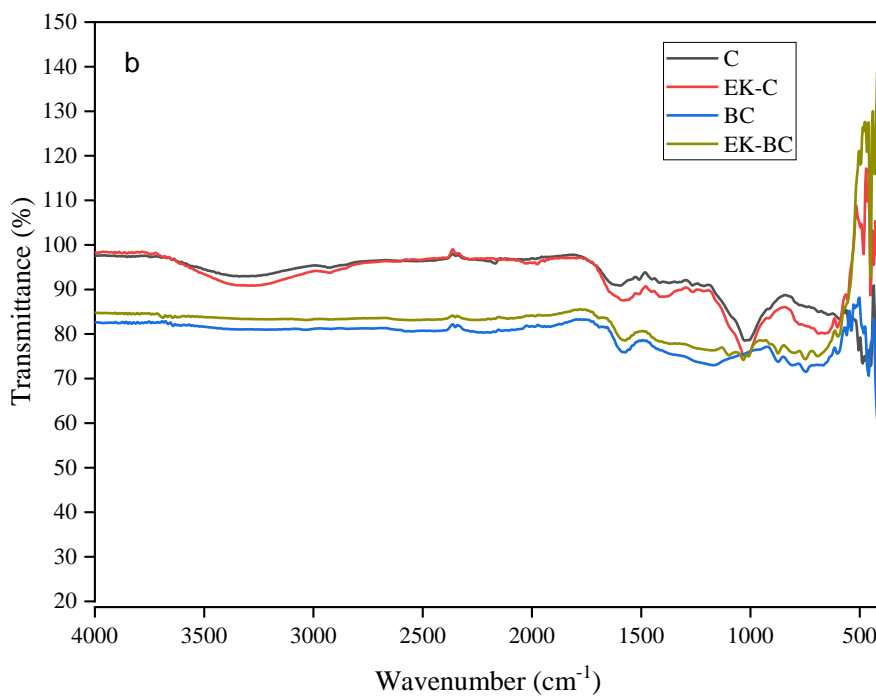
414

415 **Fig. 5.** (a) EDS analysis and spectrum of compost, (b) SEM image showing EDS analysis area,
 416 and (c) EDS map showing detection of copper (red). Scale bars: 20 μm .

417



418



419

420 **Fig. 6.** (a) Zeta potential of compost before and after EK operations; (b) FTIR spectra of
421 compost and biochar before and after EK operations.

422

423 *3.4. Performance of EK-RFM under fixed current*

424 For further enhancement of the EK process in contaminant removal, additional EK-C treatment
425 was carried out with the application of a constant electric current at 20.00 mA. Maintaining
426 electric current instead of voltage at a constant value successfully enhanced the total copper
427 removal from 45.65% to 84.09% in the EK-C treatment (**Table 3**). **20.00 mA is slightly higher**
428 **than the maximum current of 18.49 nM observed under constant voltage, but this difference is**
429 **insignificant**. The main reason was that imposing a fixed electric current increased the rate of
430 transport by accelerating the movement of the acid front. Copper concentration dramatically
431 declined from the initial concentration of 1000 mg kg⁻¹ to 41.50 mg kg⁻¹ in S1, and then slowly
432 rose to about 315.50 mg kg⁻¹ in S5 (**Fig. 7b**). The soil pH distribution, which was consistent
433 with the copper removal results (**Fig. 7b**), represented an acidic environment throughout the
434 soil, ranging between pH 2.28 and pH 3.06 (much lower than initial soil pH 5.44). The soil pH
435 in S5 was 3.06 at which copper ions will be solubilized and removed from the soil.

436 To evaluate the performance of EK-(BC+C) under fixed electric current, Exp-08 was
437 implemented under a fixed electric current (20 mA), and the results are displayed in **Fig. 7c**.
438 The total Cu removal changed from 31.01% to 48.51% in the EK-(10%BC+90%C) at the fixed
439 value of electric current (**Table 3**); however, it was not significant compared to the EK-100%C
440 experiment under the same electric current. It seems that maintaining the electric current at a
441 fixed level could not lower the pH in S5, which caused the copper accumulation in that section,
442 although it was effective in lowering soil pH in S1-S4. The reason may be the presence of the
443 biochar in the RFM mixture, which could slightly reduce the electric conductivity of the soil
444 and increase soil pH near the cathode (S5) (**Fig. 7**). It would reduce the impact of humic
445 substances of compost in lowering soil pH and binding metal ions from the contaminated soil.

446 Additionally, the average voltage during EK-(10%BC+90%C) treatment was 24.62 V, which
447 was higher than that during EK-(100%C) at 20.45 V, suggesting lower power consumption
448 using compost RFM.

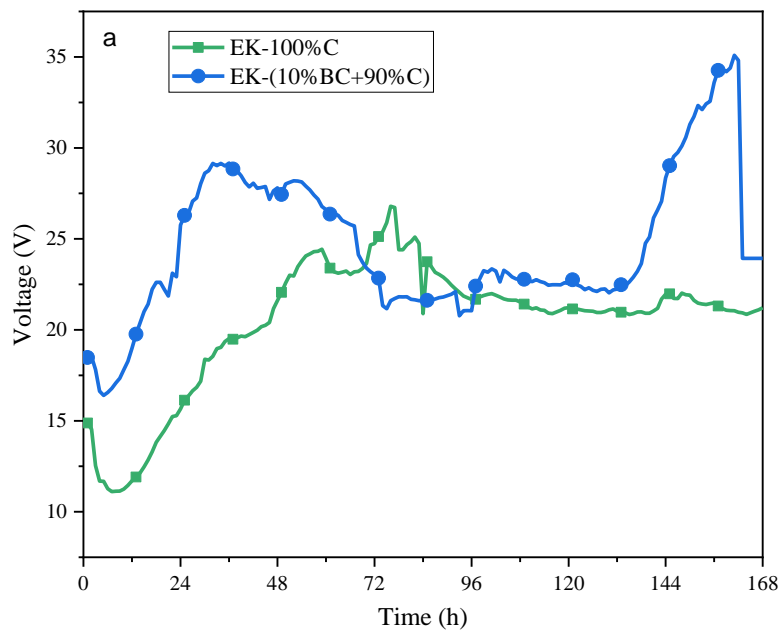
449 Considering the remediation efficiency, energy consumption per unit volume E_u (Wh m⁻³)
450 was calculated according to Eq. (3) [43]:

$$451 \quad E_u = \frac{1}{V_s} \int V I dt \quad (3)$$

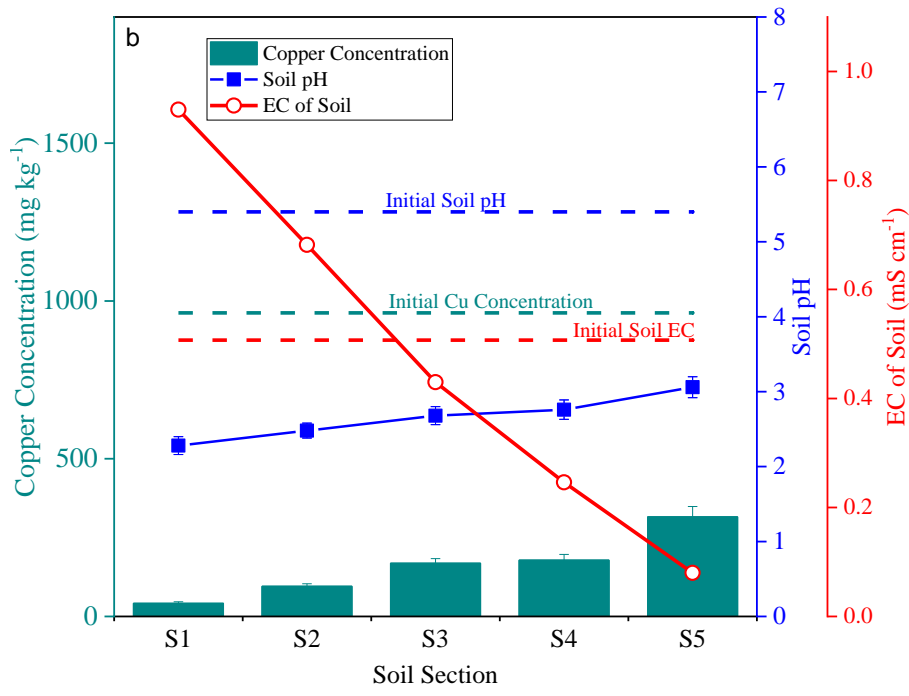
452 where V is the applied voltage (V), I the electric current (A), t the operation time (h) and V_s the
453 volume of soil treated (m³). As shown in Fig. 8, EK-(BC+C) operations generated less total
454 copper removal than EK-100%C; however, offering higher total Cu removal compared to the
455 conventional EK test. The EK remediation treatments, either with or without compost RFM,
456 consumed almost similar electrical energy, while the total copper removal in the EK
457 remediation coupled with compost RFM was nearly 45 times that in conventional EK
458 remediation (Table 3). The application of constant electric current in the EK-C treatment
459 outstandingly improved the total copper removal to 84.09% compared with that under constant
460 voltage (45.65%), although at the same dramatically increased the energy consumption from
461 10.90 kWh m⁻³ to 39.07 kWh m⁻³. With renewable energy, especially solar power under rapid
462 research and development, the practicability of this technique is expected to be extended. The
463 application of EK-(BC+C) under a constant current would be limited due to its high-energy
464 consumption demand as well as no significant improvement in removal rate in comparison with
465 the EK-C under the same electric current.

466 Mass balance and total extraction of copper after the EK operations were calculated and
467 presented in Table 3. All experiments demonstrated excellent mass balance (95.89%-
468 103.63%). The results indicated the success of different EK operations for copper removal,
469 whether maintaining a constant electric current or a constant electric voltage.

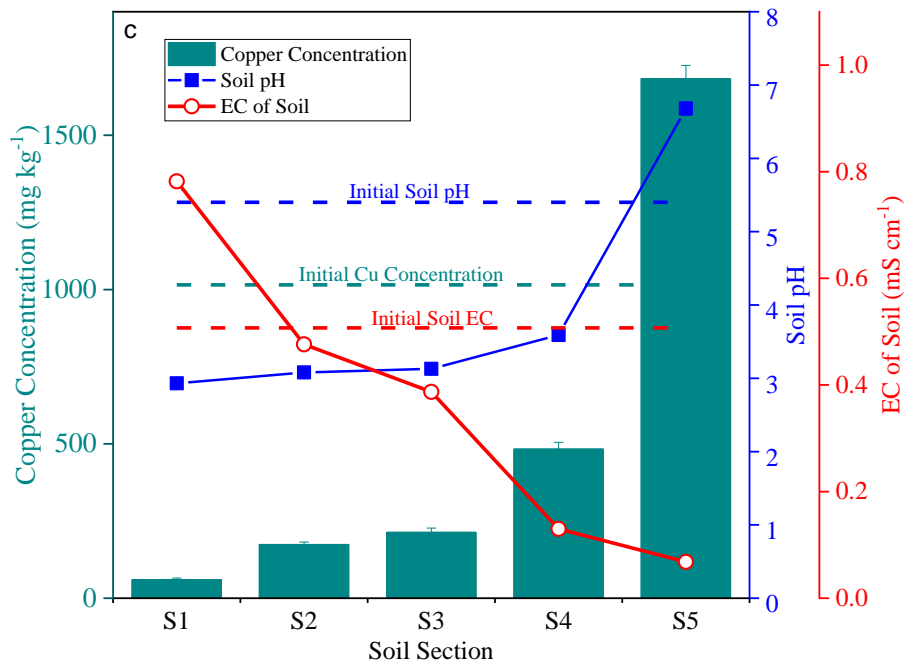
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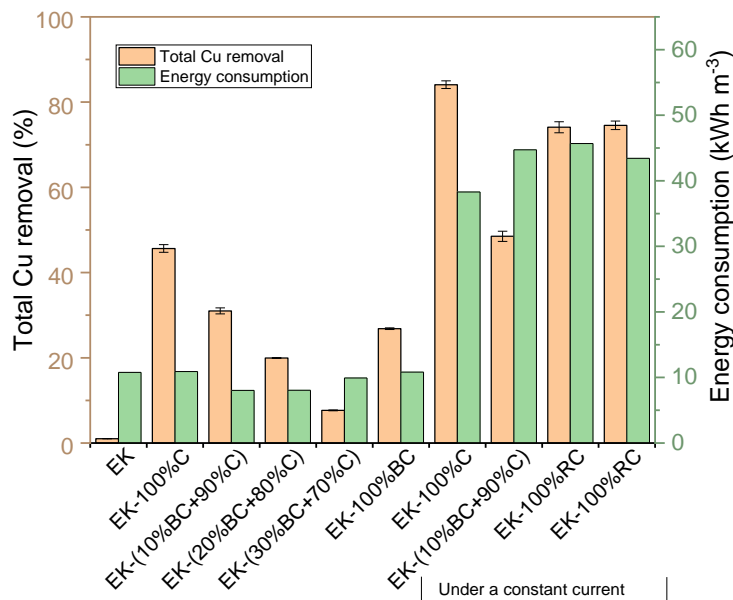
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473

474 **Fig. 7.** (a) Variation of voltage over time for EK-100%C and EK-(10%BC+90%C) treatments
 475 under a constant current; Copper concentration, pH and electric conductivity in soil sections
 476 after (b) EK-100%C and (c) EK-(10% BC+90%) treatment under a constant current.

477



478

479 **Fig. 8.** Comparison of total copper removal and associated power consumption per unit volume
 480 of soil during different EK treatment.

481

482 **Table 3.** Mass balance and total copper removal in the EK tests.

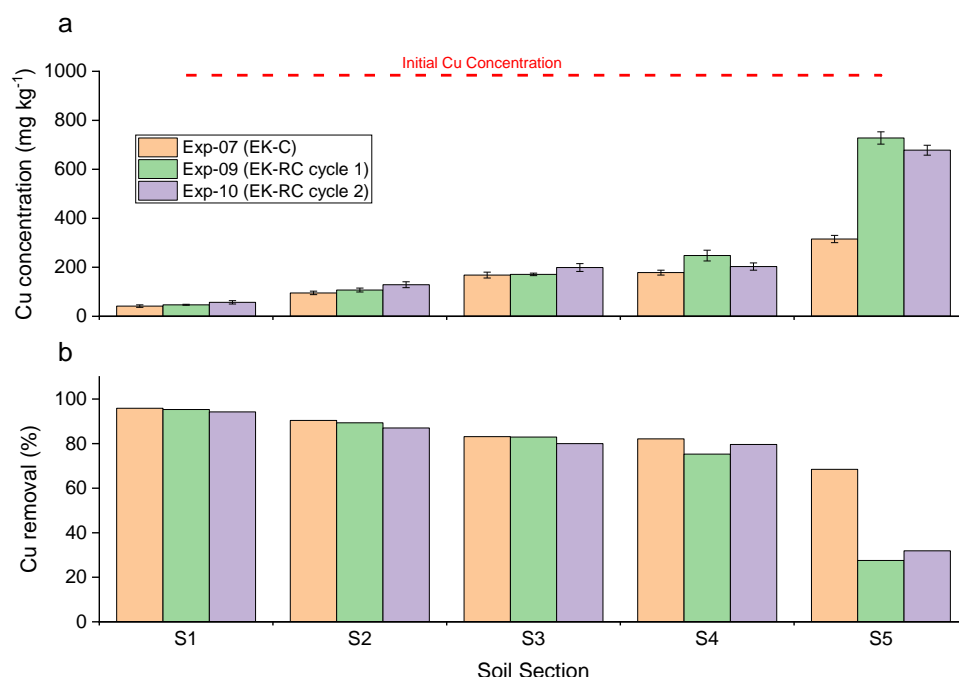
Exp. no.	Initial Cu (mg)	Residual soil Cu (mg)	Cu in RFM (mg)	Electrolyte/pore water Cu (mg)	Mass balance (%)	Total removal of Cu (%)
Exp-01	1010	999.60	N/A	8	99.76	1.03±0.02
Exp-02	980	532.60	405.30	10	96.72	45.65±0.90
Exp-03	1020	703.60	353.50	0	103.63	31.01±0.70
Exp-04	1025	820.60	213.50	3	101.18	19.94±0.11
Exp-05	1025	946.40	105	0	102.57	7.67±0.09
Exp-06	999	731	220	7	95.89	26.83±0.20
Exp-07	1005	159.90	840	0	99.49	84.09±0.90
Exp-08	1015	522.60	455	0	96.31	48.51±1.20
Exp-09	1005	260.20	730	8	99.32	74.11±1.30
Exp-10	995	253.20	737	3	99.82	74.55±1.01

483

484 *3.6. Regeneration of compost RFM and its reuse*

485 Regeneration of RFM is of importance as it can reduce the EK operation cost and minimize
486 further waste management problems. Furthermore, it would be interesting to note that such
487 regenerated RFM can be further used in the EK-RFM system for different cycles. In this study,
488 compost RFM was regenerated by acid extraction, with approximately 85% of Cu in RFM being
489 removed. In addition, about 35 g of compost remained after the regeneration process, with a
490 mass loss of 50%. The compost was recycled twice after being used in the EK-RFM system for
491 copper removal to evaluate its suitability for reuse. Since compost lost about 50% of its weight
492 during each regeneration process as well as analysis purposes, an additional fresh compost was
493 adequately mixed with the recycled compost to get a total of 70 g before reuse. An additional
494 EK-C experiment by maintaining the electric current at the fixed value (20.00 mA) (Exp-09)
495 was carried out using the recycled compost. After the EK process, the soil was removed from

496 the cell for analysis, and the RFM was taken out for regeneration purposes. The same procedure
 497 was performed to extract copper from the RFM and then reused in another EK-C test (Exp-10).
 498 The experimental results revealed that the total Cu removal rate decreased by nearly 10% (from
 499 84.09% to 74.11%) in both experiments (**Table 3**). The copper removal efficiency reduced
 500 slightly in S4 and considerably in S5 (**Fig. 9**). The reason may be that the compost likely lost
 501 the humic substances to some extent when extracted with the acid solution, which affected the
 502 performance of compost RFM in copper sorption during the EK treatment. Nevertheless,
 503 regenerated compost is still effective, showing high efficiency of copper removal compared to
 504 conventional EK process.



505
 506 **Fig. 9.** (a) Cu concentration and (b) Cu removal efficiency across the soil sections after the EK
 507 experiments coupled with the recycled compost RFM.

508

509 4. Conclusions

510 This study evaluated EK remediation of copper-contaminated kaolinite soil in conjunction with
 511 compost or compost-biochar mixture as novel RFMs. In the EK tests under a constant voltage,
 512 the total removal of copper increased from 1.03% in the EK experiment to 45.65% in the
 513 compost RFM-EK experiment. The key advantages of compost RFM-EK compared to other

514 EK processes include its great capacity in buffering the alkaline soil pH near the cathode zone
515 where the high copper accumulation occurred, eliminating the need for chemical agents which
516 may cause damage to the soil environment and increase the treatment cost. The results
517 demonstrated that mixing biochar with compost in the RFM did not improve the removal of
518 copper ions in the soil during the EK-RFM operation. The total removal of copper ions by the
519 EK treatment decreased in the order of EK-100%C > EK-(10%BC+90%C) > EK-
520 (20%BC+80%C) > EK-(30%BC+70%C) > EK. The application of a constant electric current
521 in the EK-100%C treatment further improved copper extraction in the soil to 84.09%, although
522 simultaneously increasing the energy consumption. Furthermore, compost was regenerated and
523 reused in the EK process, although the total Cu removal decreased from 84.09% to 74.11%.
524 The findings confirm compost as a promising green RFM which enhances heavy metals
525 removal in the EK process, especially under constant electric current, by generating a low pH
526 environment through the soil.

527

528 **Acknowledgements**

529 We thank the University of Technology Sydney for a PhD studentship, and a grant from the
530 111 Project (D18012), Ministry of Education, China.

531

532 **References**

- 533 [1] L. Yuan, X. Xu, H. Li, N. Wang, N. Guo, H. Yu, Development of novel assisting
534 agents for the electrokinetic remediation of heavy metal-contaminated kaolin,
535 *Electrochim. Acta.* 218 (2016) 140–148.
- 536 [2] Y. Liu, J. Chen, Z. Cai, R. Chen, Q. Sun, M. Sun, Removal of copper and nickel from
537 municipal sludge using an improved electrokinetic process, *Chem. Eng. J.* 307 (2016)
538 1008–1016.
- 539 [3] H. Frick, S. Tardif, E. Kandeler, P.E. Holm, K.K. Brandt, Assessment of biochar and

- 540 zero-valent iron for in-situ remediation of chromated copper arsenate contaminated
541 soil, *Sci. Total Environ.* 655 (2019) 414–422.
- 542 [4] M. Fagnano, D. Agrelli, A. Pascale, P. Adamo, N. Fiorentino, C. Rocco, O. Pepe, V.
543 Ventorino, Copper accumulation in agricultural soils: Risks for the food chain and soil
544 microbial populations, *Sci. Total Environ.* 734 (2020) 139434.
- 545 [5] R. Iannelli, M. Masi, A. Ceccarini, M.B. Ostuni, R. Lageman, A. Muntoni, D. Spiga,
546 A. Poletini, A. Marini, R. Pomi, Electrokinetic remediation of metal-polluted marine
547 sediments: Experimental investigation for plant design, *Electrochim. Acta.* 181 (2015)
548 146–159.
- 549 [6] I. Hassan, E. Mohamedelhasan, E.K. Yanful, Solar powered electrokinetic
550 remediation of Cu polluted soil using a novel anode configuration, *Electrochim. Acta.*
551 181 (2015) 58–67.
- 552 [7] L. Yuan, X. Xu, H. Li, Q. Wang, N. Wang, H. Yu, The influence of macroelements on
553 energy consumption during periodic power electrokinetic remediation of heavy metals
554 contaminated black soil, *Electrochim. Acta.* 235 (2017) 604–612.
- 555 [8] Z. Sun, B. Wu, P. Guo, S. Wang, S. Guo, Enhanced electrokinetic remediation and
556 simulation of cadmium- contaminated soil by superimposed electric field, 233 (2019)
557 17–24.
- 558 [9] R. López-Vizcaíno, A. Yustres, M.J. León, C. Saez, P. Cañizares, M.A. Rodrigo, V.
559 Navarro, Multiphysics Implementation of Electrokinetic Remediation Models for
560 Natural Soils and Porewaters, *Electrochim. Acta.* 225 (2017) 93–104.
- 561 [10] C. Peppicelli, P. Cleall, D. Sapsford, M. Harbottle, Changes in metal speciation and
562 mobility during electrokinetic treatment of industrial wastes: Implications for
563 remediation and resource recovery, *Sci. Total Environ.* 624 (2018) 1488–1503.
- 564 [11] A.T. Yeung, Milestone developments, myths, and future directions of electrokinetic
565 remediation, *Sep. Purif. Technol.* 79 (2011) 124–132.

- 566 [12] S.S. Al-Shahrani, E.P.L. Roberts, Electrokinetic removal of caesium from kaolin, J.
567 Hazard. Mater. 122 (2005) 91–101.
- 568 [13] A. Kaya, Y. Yukselen, Zeta potential of soils with surfactants and its relevance to
569 electrokinetic remediation, 120 (2005) 119–126.
- 570 [14] J.N. Hahladakis, N. Lekkas, A. Smpontas, E. Gidarakos, Sequential application of
571 chelating agents and innovative surfactants for the enhanced electroremediation of real
572 sediments from toxic metals and PAHs, Chemosphere. 105 (2014) 44–52.
- 573 [15] T. Suzuki, M. Moribe, Y. Okabe, M. Niinae, A mechanistic study of arsenate removal
574 from artificially contaminated clay soils by electrokinetic remediation, J. Hazard.
575 Mater. 254–255 (2013) 310–317.
- 576 [16] M. Bahemmat, M. Farahbakhsh, M. Kianirad, Humic substances-enhanced
577 electroremediation of heavy metals contaminated soil, J. Hazard. Mater. 312 (2016)
578 307–318.
- 579 [17] R. Fu, D. Wen, X. Xia, W. Zhang, Y. Gu, Electrokinetic remediation of chromium
580 (Cr)-contaminated soil with citric acid (CA) and polyaspartic acid (PASP) as
581 electrolytes, Chem. Eng. J. 316 (2017) 601–608.
- 582 [18] M.W. Lim, E. Von Lau, P.E. Poh, A comprehensive guide of remediation technologies
583 for oil contaminated soil — Present works and future directions, Mar. Pollut. Bull. 109
584 (2016) 14–45.
- 585 [19] Y. Tian, C. Boulangé-Lecomte, A. Benamar, N. Giusti-Petrucciani, A. Duflot, S.
586 Olivier, C. Frederick, J. Forget-Leray, F. Portet-Koltalo, Application of a crustacean
587 bioassay to evaluate a multi-contaminated (metal, PAH, PCB) harbor sediment before
588 and after electrokinetic remediation using eco-friendly enhancing agents, Sci. Total
589 Environ. 607–608 (2017) 944–953.
- 590 [20] L. Cang, Q.Y. Wang, D.M. Zhou, H. Xu, Effects of electrokinetic-assisted
591 phytoremediation of a multiple-metal contaminated soil on soil metal bioavailability

- 592 and uptake by Indian mustard, *Sep. Purif. Technol.* 79 (2011) 246–253.
- 593 [21] R. Bi, M. Schlaak, E. Siefert, R. Lord, H. Connolly, Influence of electrical fields (AC
594 and DC) on phytoremediation of metal polluted soils with rapeseed (*Brassica napus*)
595 and tobacco (*Nicotiana tabacum*), *Chemosphere.* 83 (2011) 318–326.
- 596 [22] K.Y. Lee, K.W. Kim, Heavy metal removal from shooting range soil by hybrid
597 electrokinetics with bacteria and enhancing agents, *Environ. Sci. Technol.* 44 (2010)
598 9482–9487.
- 599 [23] C. Yuan, T.S. Chiang, The mechanisms of arsenic removal from soil by electrokinetic
600 process coupled with iron permeable reaction barrier, *Chemosphere.* 67 (2007) 1533–
601 1542.
- 602 [24] F. Xue, Y. Yan, M. Xia, F. Muhammad, L. Yu, F. Xu, Y. Shiao, D. Li, B. Jiao, Electro-
603 kinetic remediation of chromium-contaminated soil by a three-dimensional electrode
604 coupled with a permeable reactive barrier, *RSC Adv.* 7 (2017) 54797–54805.
- 605 [25] R. Ghobadi, A. Altaee, J.L. Zhou, P. McLean, S. Yadav, Copper removal from
606 contaminated soil through electrokinetic process with reactive filter media,
607 *Chemosphere.* 252 (2020) 126607.
- 608 [26] J. Wei, F. Yuan, Z. Hong, H. Wang, Simultaneous removal of 2, 4-dichlorophenol and
609 Cd from soils by electrokinetic remediation combined with activated bamboo charcoal,
610 *Chemosphere.* 176 (2010) 715–720.
- 611 [27] L. Ren, H. Lu, L. He, Y. Zhang, Enhanced electrokinetic technologies with
612 oxidization-reduction for organically-contaminated soil remediation, *Chem. Eng. J.* 247
613 (2014) 111–124.
- 614 [28] J.H. Chang, S.F. Cheng, The remediation performance of a specific electrokinetics
615 integrated with zero-valent metals for perchloroethylene contaminated soils, *J. Hazard.*
616 *Mater.* 131 (2006) 153–162.
- 617 [29] M. Farrell, D.L. Jones, Use of composts in the remediation of heavy metal

- 618 contaminated soil, *J. Hazard. Mater.* 175 (2010) 575–582.
- 619 [30] S. Tandy, J.R. Healey, M.A. Nason, J.C. Williamson, D.L. Jones, Remediation of metal
620 polluted mine soil with compost: Co-composting versus incorporation, *Environ. Pollut.*
621 157 (2009) 690–697.
- 622 [31] J. Hermana, E. Nurhayati, Removal of Cr³⁺ and Hg²⁺ using compost derived from
623 municipal solid waste, *Sustain. Environ. Res.* 20 (2010) 257–261.
- 624 [32] R. Clemente, M.P. Bernal, Fractionation of heavy metals and distribution of organic
625 carbon in two contaminated soils amended with humic acids, *Chemosphere.* 64 (2006)
626 1264–1273.
- 627 [33] L. Beesley, E. Moreno-jiménez, J.L. Gomez-eyles, Effects of biochar and greenwaste
628 compost amendments on mobility , bioavailability and toxicity of inorganic and
629 organic contaminants in a multi-element polluted soil, *Environ. Pollut. J.* 158 (2010)
630 2282–2287.
- 631 [34] H.A. Alhashimi, C.B. Aktas, Life cycle environmental and economic performance of
632 biochar compared with activated carbon: A meta-analysis, *Resour. Conserv. Recycl.*
633 118 (2017) 13–26.
- 634 [35] A. Altaee, R. Smith, S. Mikhalovsky, The feasibility of decontamination of reduced
635 saline sediments from copper using the electrokinetic process, *J. Environ. Manage.* 88
636 (2008) 1611–1618.
- 637 [36] C. Parsons, E. Margui, E. Pili, G.H. Floor, G. Roman-ross, L. Charlet, Quantification
638 of trace arsenic in soils by field-portable X-ray fluorescence spectrometry :
639 Considerations for sample preparation and measurement conditions, *J. Hazard. Mater.*
640 262 (2013) 1213–1222.
- 641 [37] T. Radu, D. Diamond, Comparison of soil pollution concentrations determined using
642 AAS and portable XRF techniques, *J. Hazard. Mater.* 171 (2009) 1168–1171.
- 643 [38] C. Kilbride, J. Poole, T.R. Hutchings, A comparison of Cu, Pb, As, Cd, Zn, Fe, Ni and

- 644 Mn determined by acid extraction/ICP-OES and ex situ field portable X-ray
645 fluorescence analyses, *Environ. Pollut.* 143 (2006) 16–23.
- 646 [39] M. Rouillon, M.P. Taylor, Can field portable X-ray fluorescence (pXRF) produce high
647 quality data for application in environmental contamination research?, *Environ. Pollut.*
648 214 (2016) 255–264.
- 649 [40] M. Rouillon, P.J. Harvey, L.J. Kristensen, S.G. George, M.P. Taylor, VegeSafe: A
650 community science program measuring soil-metal contamination, evaluating risk and
651 providing advice for safe gardening, *Environ. Pollut.* 222 (2017) 557–566.
- 652 [41] M.T. Alcántara, J. Gómez, M. Pazos, M.A. Sanromán, Electrokinetic remediation of
653 lead and phenanthrene polluted soils, *Geoderma.* 173–174 (2012) 128–133.
- 654 [42] M. Zhou, J. Xu, S. Zhu, Y. Wang, H. Gao, Exchange electrode-electrokinetic
655 remediation of Cr- contaminated soil using solar energy, *Sep. Purif. Technol.* 190
656 (2017) 297–306.
- 657 [43] J.N. Hahladakis, A. Latsos, E. Gidarakos, Performance of electroremediation in real
658 contaminated sediments using a big cell, periodic voltage and innovative surfactants, *J.*
659 *Hazard. Mater.* 320 (2016) 376–385.
- 660 [44] A. Figueroa, C. Cameselle, S. Gouveia, H.K. Hansen, Electrokinetic treatment of an
661 agricultural soil contaminated with heavy metals, *J. Environ. Sci. Heal. - Part A*
662 *Toxic/Hazardous Subst. Environ. Eng.* 51 (2016) 691–700.
- 663 [45] A.T. Yeung, Y.Y. Gu, A review on techniques to enhance electrochemical remediation
664 of contaminated soils, *J. Hazard. Mater.* 195 (2011) 11–29.
- 665 [46] C. Cameselle, R.A. Chirakkara, K.R. Reddy, Electrokinetic-enhanced
666 phytoremediation of soils: Status and opportunities, *Chemosphere.* 93 (2013) 626–636.
- 667 [47] N. Habibul, Y. Hu, G.-P. Sheng, Microbial fuel cell driving electrokinetic remediation
668 of toxic metal contaminated soils, *J. Hazard. Mater.* 318 (2016) 9–14.
- 669 [48] M. Masi, A. Ceccarini, R. Iannelli, Multispecies reactive transport modelling of

- 670 electrokinetic remediation of harbour sediments, *J. Hazard. Mater.* 326 (2017) 187–
671 196.
- 672 [49] M. Pazos, M.A. Sanromán, C. Cameselle, Improvement in electrokinetic remediation
673 of heavy metal spiked kaolin with the polarity exchange technique, *Chemosphere.* 62
674 (2006) 817–822.
- 675 [50] J.S. Yang, M.J. Kwon, J. Choi, K. Baek, E.J. O’Loughlin, The transport behavior of
676 As, Cu, Pb, and Zn during electrokinetic remediation of a contaminated soil using
677 electrolyte conditioning, *Chemosphere.* 117 (2014) 79–86.
- 678 [51] M. Rezaee, G. Asadollahfardi, C. Gomez-Lahoz, M. Villen-Guzman, J.M. Paz-Garcia,
679 Modeling of electrokinetic remediation of Cd- and Pb-contaminated kaolinite, *J.*
680 *Hazard. Mater.* 366 (2019) 630–635.
- 681 [52] R. Ortiz-soto, H.K. Hansen, D. Leal, C. Gutierrez, A. Aracena, A. Rojo, Electrokinetic
682 remediation of manganese and zinc in copper mine tailings, *J. Hazard. Mater.* 365
683 (2019) 905–911.
- 684 [53] G. Hotová, V. Slovák, T. Zelenka, R. Maršálek, A. Parchanská, The role of the oxygen
685 functional groups in adsorption of copper (II) on carbon surface, *Sci. Total Environ.*
686 711 (2020) 135436.
- 687 [54] G. Zeng, H. Wu, J. Liang, S. Guo, L. Huang, P. Xu, Y. Liu, Y. Yuan, X. He, Y. He,
688 Efficiency of biochar and compost (or composting) combined amendments for
689 reducing Cd, Cu, Zn and Pb bioavailability, mobility and ecological risk in wetland
690 soil, *RSC Adv.* 5 (2015) 34541–34548.
- 691