

Electrocoagulation of food waste digestate and the suitability of recovered solids for application to agricultural land.

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1 **Title:** Electrocoagulation of food waste digestate and the suitability of recovered solids for
2 application to agricultural land

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15 **Abstract**

16 Digestates from anaerobic digestion (AD) of food waste contain fertiliser nutrients (such as P
17 and N) which are valuable for agricultural purposes and can be environmentally hazardous if
18 disposal is uncontrolled. Here, we applied electrocoagulation (EC) for treatment of digestates,
19 to separate liquids and nutrient-rich solids. Coagulant-dosing electrocoagulation (CDEC) was
20 used to compare Al and steel anodes for treatment of digestate from AD fermenters fed a
21 controlled diet representative of food waste. When applying metal dosing concentrations of
22 0 - 4.66 mM, Fe was found to be up to 29.8% superior to Al in terms of aiding removal of
23 chemical oxygen demand (COD). To mitigate plate fouling, the digestate was diluted to 0.25x

24 and 0.1x concentrations to enable successful treatment by continuous-flow
25 electrocoagulation (CFEC). The highest recovery of soluble P per Fe added by CFEC was 22.4
26 mg-P g-Fe⁻¹. This was achieved when using 2.33 mM Fe to treat a 0.25x dilution of digestate.
27 In comparison to a control, these optimal conditions also caused a reduction of 4.5 mg L⁻¹ (or
28 87.7%) of soluble P and the removal of 254 mg L⁻¹ (33.2%) of COD in the filtrate. The NH₄⁺
29 concentration in filtrate was not influenced by EC treatment. Analyses of a range of known
30 toxic elements (Cu, Ni, Zn, As, Cd, Cr, Mo and Pb) indicates that the solids recovered by CFEC
31 could be suitable for application to land.

32 **Keywords:** Anaerobic digestion, Electrocoagulation, Nutrient recovery, Phosphorous,
33 Nitrogen.

34 **1. Introduction**

35 The use of food waste as feedstock for the production of biomethane by industrial anaerobic
36 digestion (AD) has grown significantly over the last decade (Moult et al., 2018). In addition to
37 gaseous fuel production, a further product of AD are slurries (or digestates) which contain
38 residual bio-solid material. Digestates derived from the AD of food waste often contain
39 nutrients such as nitrogen (N) and phosphorous (P) which are conventionally sourced from
40 energy intensive fertiliser production methods for their use in agriculture (Tampio et al.,
41 2016). Therefore, the recovery of energy and nutrients from food waste was recently
42 described as economically important and essential for the sustainable development of human
43 society (Xu et al., 2018).

44 Although food waste digestate is rich in valuable fertiliser nutrients, its application in
45 agriculture remains challenging. The nutrients in raw digestates are contained in large
46 amounts of water. The overall volume of water not only has negative implications for the

47 costs associated with digestate transportation but also increases the possibility of
48 environmental damage via eutrophication or contamination of natural water systems
49 (Tampio et al., 2016). To enable targeted fertiliser application and limit the environmental
50 risk, suitable digestate treatment methods are necessary to extract fertiliser nutrients (such
51 as N, P and K) out of solution. Typically, solid and liquid separation methodologies initially use
52 a solids thickening technology (which often consists of settling, filtration or centrifugation)
53 and incorporates the use of a polyelectrolyte or metal chloride coagulants (e.g. ferric chloride)
54 (Monfet et al., 2018). The thickening of solids in digestate is usually followed by a dewatering
55 step, which is commonly carried out by using either a filter press or centrifugation (Knocke et
56 al., 1993; Wakeman, 2007).

57 Addition of coagulant during separation of solids and liquids is beneficial because solids can
58 be bound together (or coagulated) to increase floc size (Knocke et al., 1993). This increase in
59 floc size facilitates the use of lower-cost dewatering technology by enabling the use of larger
60 porosity filters and reduced filtration pressures (Wakeman, 2007). An analogous alternative
61 to the use of polyelectrolyte or metal chloride coagulants for digestate treatment is the
62 application of electrocoagulation (EC) (Al-Qodah et al., 2019). During EC, the dissolution of a
63 sacrificial metal anode is driven electrochemically (corrosion) causing the release of metal ions into
64 the digestate according to the following equations:

65 At the cathode:



67 At the anode:



70 The amount of metal ions released from the anode (w) into the waste stream can be derived
71 by using Faraday's law:

$$72 \quad w = (I t M)/(z F) \quad (4)$$

73 Where I is the current (A), t is the process time (s), M is the molar mass of the electrode metal
74 (g mol^{-1}), z is the valence of the anode metal and F is Faraday's constant ($96,485 \text{ C mol}^{-1}$).

75 Once in the digestate, the metal ions react with OH^- groups to form hydroxides which can
76 coagulate solids by complexation or electrostatic attraction (Hakizimana et al., 2017).

77 Electrodes are most often constructed out of Fe and Al because these metals have high
78 valence, are cost effective and widely available (Hakizimana et al., 2017; Tirado et al., 2018).

79 Furthermore, the respective hydroxides (e.g. $\text{Al}(\text{OH})_3$ and $\text{Fe}(\text{OH})_2$ are highly insoluble (K_{sp}
80 3×10^{-34} and 8×10^{-16} , respectively). Multiple factors influence the cost and performance of EC

81 treatment such as the metal ion concentration (Mameri et al., 1998), initial pH (Tezcan Un et
82 al., 2018), retention time (Tezcan Un et al., 2018), electrode material (Devlin et al., 2019), EC

83 chamber design (Kabdaşlı et al., 2012) and salt concentration (Yıldız et al., 2008).

84 EC technology can be operated in various treatment configurations which include 'coagulant
85 dosing' (where the electrodes are used to generate a concentrated solution of coagulant

86 which is then introduced into the effluent stream), 'batch' (where the electrodes are corroded
87 directly into the effluent but treatment is carried out by processing discrete volumes of

88 effluent at a time) (Tezcan Ün et al., 2006; Yıldız et al., 2008) and 'continuous flow' (where
89 electrodes are operating continuously and in direct contact with the effluent as it flows

90 through the reactor/cell) (Benazzi et al., 2015; Makwana and Ahammed, 2016). Each
91 configuration has specific advantages. Coagulant dosing limits direct contact of digestate with

92 plates and mitigates electrode fouling. Batch treatment is carried out by submersing the

93 anode and cathode into the digestate and requires minimal equipment. Continuous flow EC
94 enables uninterrupted processing of large treatment loads (Benazzi et al., 2015). Previously,
95 EC has been successfully applied to treat a range effluent streams including those from dairies
96 and slaughterhouses (Reilly et al., 2019), the textile industry (Alinsafi et al., 2005), landfill
97 leachates (Li et al., 2011) and wastewater (Kuokkanen et al., 2013). EC is potentially
98 advantageous over conventional coagulation methods for digestate treatment as it excludes
99 the addition of synthetic polymers to digestate, which minimises the potential for
100 contamination of land during spreading of the recovered solids onto agricultural soils
101 (Lapointe and Barbeau, 2019). Furthermore, metal electrodes occupy less space than their
102 corresponding metal chloride coagulants. The reduced footprint is beneficial for both
103 transportation and storage requirements. Additionally, storage of electrodes is significantly
104 safer than that of large volumes of metal chloride solutions.

105 A limited number of studies have successfully investigated the use of EC to treat digestates
106 from AD. To date, these studies have mainly focused on effluents from AD plants processing
107 sewage and municipal solid waste (MSW) (Fernandes et al., 2017; Huang et al., 2017;
108 Makwana and Ahammed, 2016; Olvera-Vargas et al., 2019). For example Huang et al. (2017)
109 demonstrated that the use of Fe^{2+} EC to recover phosphate from anaerobic sludge worked
110 optimally between pH 6-8 and concluded that the recovery of phosphate by EC using
111 electrodes made of Fe was more efficient than using those made of Al. Makwana and
112 Ahammed (2017) and Fernandes et al., (2017) reported that chemical oxygen demand (COD)
113 removal of 71% and 80% could be achieved from digestates derived from urban wastewater
114 and MSW, respectively. Makwana and Ahammed (2016) described a continuous process
115 which they applied to effluent from an upflow anaerobic sludge blanket (UASB) reactor
116 treating municipal wastewater. When using a current density of 20 mA cm^{-2} the process

117 removed 67.2% COD and reduced initial phosphate concentrations (ranging between 4.5-6.4
118 mg L⁻¹) down to 0.57 mg L⁻¹.

119 However, there is little information in current literature regarding the application of EC to
120 process digestate from anaerobic digesters fed food waste. Furthermore, the majority of EC
121 studies have demonstrated batch operation. Liu et al. (2015) successfully performed EC on
122 the effluent from co-digestion of dairy manure and food waste, although that particular study
123 did not include solids dewatering and the analysis focused on the recovered liquids. Hence,
124 the objective of this study was to use continuous EC to coagulate digestate from food waste
125 AD. This study included a comparison between using Fe and Al as electrodes, with solid and
126 liquid fractions being chemically analysed following a post-EC dewatering stage. Here, we also
127 report the concentrations of a range of potentially toxic elements (PTE) in the EC-treated
128 digestate and for the first time, consider their implications on the suitability of the recovered
129 solids for use as a fertiliser on agricultural soils.

130 **2. Materials & Methods**

131 **2.1 Digestate preparation and characteristics**

132 The digestate used in this study was sourced from laboratory-scale anaerobic digesters, which
133 were operated in duplicate. The anaerobic digesters were inoculated with sludge digestate
134 sourced from a mesophilic digester operating at 35°C on Monkmoor sewage treatment works
135 (Shrewsbury, UK). Each laboratory-scale digester was fed a synthetic recipe representing food
136 waste as per Jobling-Purser (2015). Feedstock was prepared weekly and stored at 5°C. Each
137 laboratory anaerobic digester was 26L and had a working volume of 23L. The digesters were
138 fed daily at a rate of 2.5 Kg-VS m⁻³ d⁻¹ of synthetic food waste and using a hydraulic retention

139 time (HRT) of 20 days. The digesters were mesophilic (35°C) and stirred at 100 rpm; they were
 140 operated for 3 HRT to ensure steady-state operation before digestate was used in EC trials.
 141 Digestate was collected each day, pooled and refrigerated (at 5°C) for no more than 7 days
 142 prior before being passed through a 500 µm sieve and used in EC trials. The characteristics of
 143 the digestate prior to EC are shown in Table 1.

Table 1. Characteristics of sieved digestate

Component	Unit	Mean ± standard deviation	
Total Solids (TS)	% (w.w.)	1.36	± 0.01
Volatile Solids (VS)	% (d.w.)	79.30	± 0.47
Ash Content	% (d.w.)	20.70	± 0.41
Total Suspended Solids (TSS)	g L ⁻¹	12.30	± 0.64
Conductivity	mS cm ⁻¹	5.23	± 0.66
Turbidity	NTU	7,545	± 785
pH		7.36	± 0.12
Total Alkalinity	mg-CaCO ₃ L ⁻¹	650	± 105
COD	mg L ⁻¹	12,250	± 433
C (Total)	% (d.w.)	46.4	± 0.5
N (Total)	% (d.w.)	8.94	± 0.03
P (Filter Solids)	% (d.w.)	0.735	± 0.111
P (Soluble)	mg L ⁻¹	21.8	± 1.1
S (Total)	% (d.w.)	0.513	± 0.066
NH ₄ -N (soluble)	mg L ⁻¹	383	± 4
NO ₃ -N (soluble)	mg L ⁻¹	20.6	± 3.1

144

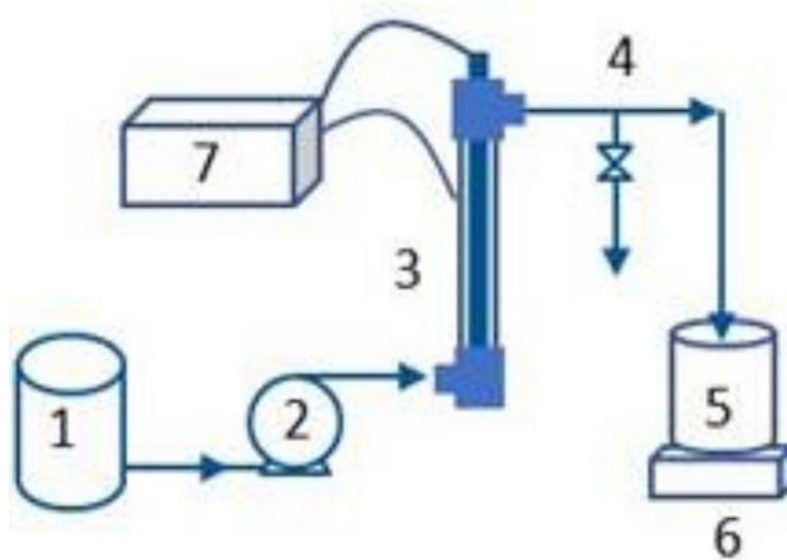
145 **2.2 Electrocoagulation processing**

146 **2.2.1 Electrocoagulation equipment**

147 Electrochemical experiments were performed using a benchtop EC system (Elentec Ltd., UK).
 148 The system included a circulation pump (Model 630S, Watson Marlow Ltd., UK) and power
 149 supply (EA Elektro-Automatik EA-PS 2042-10B, RS Components Ltd., UK). A simplified flow
 150 diagram is presented in Figure 1. The EC cell consisted of a tubular cathode in stainless steel
 151 (SS 304, ID 19 mm, length 25.8 cm) fitted with ½" BSP fittings which located the anode along

152 the middle axis of the cell. Anodic material was either mild steel (12mm diam., RS
153 Components Ltd., UK) or aluminium (6082-T6, 12mm diam., RS components). The active
154 surface area of the anode was 97.3 cm².

155 The EC equipment was operated aerobically in two electrocoagulation modes: coagulant-
156 dosing (CDEC) and continuous-flow (CFEC). Both CDEC and CFEC were operated using a
157 pumping flow rate of 20L hr⁻¹ (through the EC cell). A new anode was used for each
158 experiment.



159
160 **Figure 1. A schematic diagram of electrocoagulation equipment (not to scale). 1 = Sample**
161 **storage tank; 2 = Circulation pump; 3 = EC cell; 4 = Sampling valve; 5 = Collection beaker; 6**
162 **= Magnetic stirrer; 7 = Power supply.**

164 2.2.2 Screening experiment: Coagulant-dosing electrocoagulation (CDEC)

165 Screening experiments were completed to examine the feasibility of EC for the treatment of
166 food waste digestate and to select suitable parameters for subsequent dewatering
167 experiments. CDEC was used to create a coagulant solution by releasing Fe and Al ions from
168 the anode into solution, which was then added to digestate. This coagulant was produced as
169 follows; 17.1 mM NaCl (with a conductivity of 2 mS cm⁻¹) was pumped through the EC cell. At

170 constant flow rate, the current input was varied according to Faraday's equation (equation 4)
171 to generate concentrations of Fe or Al coagulant solutions. Either 100 ml or 250 ml of
172 digestate were made up to 1L though addition of generated electrolyte solution, resulting in
173 a 0.1 and 0.25 dilution of digestate, respectively. The final coagulant-dosing concentration of
174 Fe and Al in the diluted samples were 0.933, 1.87, 2.80, 3.73, 4.66 mM. Control samples were
175 prepared which contained the volumes of digestate described above but with 17.1 mM NaCl
176 (instead of coagulant solution). Each CDEC condition was repeated in triplicate. Therefore,
177 CDEC experimentation included a total sample size of 72 (Supplementary Table A).

178 After the contents of a CDEC beaker had been made up to 1 L (as described above), it was
179 magnetically stirred for 1 minute at 2000 rpm, prior to undergoing 30 min of paddle-mixing
180 at 40 rpm with a Stuart SW6 Flocculator (Cole-Parmer Ltd., UK). Following flocculation, the
181 samples received an undisturbed period of 15 hours at room temperature. Samples were
182 agitated at 30 rpm and 8 ml of subsample was passed through Aeropress® filter paper using
183 a handheld Aerobie press (Aeropress Inc., USA). The filtrate was used for analysis.

184 **2.2.3 Continuous-flow electrocoagulation (CFEC)**

185 Digestate was mixed with DiH₂O to achieve dilution concentrations of 0.25 or 0.1
186 (digestate:DiH₂O). The conductivity of digestate dilutions were adjusted with NaCl such that
187 all samples had a conductivity of 2 mS cm⁻¹. The diluted digestate was pumped at 20 L hr⁻¹
188 through the EC cell which was powered with 0, 2.5 and 5 amps to achieve concentrations of
189 0, 2.33, and 4.66 mM Fe from the steel anode. CFEC control (0 mM Fe) tests were carried out
190 by pumping the diluted digestate through the EC cell without an electrical current. For each
191 sample, 1L of effluent was collected in a glass beaker, which were prepared in triplicates,
192 resulting in an overall sample size of 18 for CFEC tests (Supplementary Table B). Stirring,

193 paddle-mixing and settlement of each beaker was completed as previously described (for
194 CDEC) prior to dewatering.

195 **2.3 Dewatering of CFEC samples**

196 A bespoke stainless steel filter-press system was manufactured for dewatering of CFEC
197 samples (Supplementary Figure S1). The filter-press consisted of a piston (with a holder for
198 multiple weighted plates and a lifting handle) and a cylindrical upright pipe (with an I.D. of
199 140 mm) for suspending the CFEC sample between filter papers. Samples were loaded into
200 the filter-press using a volume of 125mL or 250mL for digestate samples which had been
201 diluted with H₂O to concentrations of 0.25 or 0.1 prior to CFEC treatment, respectively. The
202 filter-press was used to apply a pressure of 48.2 kPa for 30 minutes to dewater the sample
203 and create a cake between 2x Fisher Brand QT280 (12-15µm) filter papers. Filtrate and
204 pressed cake solids were collected for analysis.

205 **2.4 Analytical Methods**

206 Total solids (TS) and volatile solids (VS) were measured gravimetrically according to the
207 standard method (APHA, 1989). Total suspended solids in samples were quantified according
208 to EPA Method 160.2 using 0.7µm Merck™ AP4004705 filters (Fisher Scientific, UK) (EPA,
209 1971). Conductivity was measured by using a Hanna HI 86304 conductivity meter equipped
210 with a HI-7632D/1 probe. A Jenway 3510 pH meter was used for pH measurements (Jenway,
211 UK). Alkalinity was quantified by using a KEM AT-710 auto-titrator (Kyoto Electronics
212 Manufacturing Co. Ltd., Japan) and the titration method of Jenkins et al. (1983). Turbidity was
213 analysed by use of a ThermoScientific Eutech TN-100 meter (Fisher Scientific, UK). COD was
214 determined using a potassium dichromate method Hach Lange LCI 400 assay kit (Hach Lange,
215 UK) and a Hach Lange DR 1900 spectrophotometer (Hach Lange, UK). Total carbon and

216 sulphur of dried samples were quantified using a Leco SC-144 BR Sulfur/Carbon Analyser
217 (LECO, USA) (Kirby et al., 2018). Total nitrogen of dried samples was determined as per AOAC
218 method 1990 968.06 by using a Leco FP528 Nitrogen/Protein Determinator (LECO, USA)
219 (Helrich, 1990). Ammonium in filtrate was measured according to AOAC method 1990 920.03
220 using a Foss Kjeltac 8400 Kjeldahl Analyser (Foss, Denmark) (Helrich, 1990). Nitrate was
221 measured using Merck Millipore MColorTest nitrate test kit and a Hach Lange DR 1900
222 spectrophotometer (Hach Lange Ltd., UK) at 520 nm. All other elements were quantified using
223 ICP-MS (Perkin Elmer NexION 2000, Perkin Elmer, USA) according to EPA Method 6020B - 2
224 (EPA, 2014).

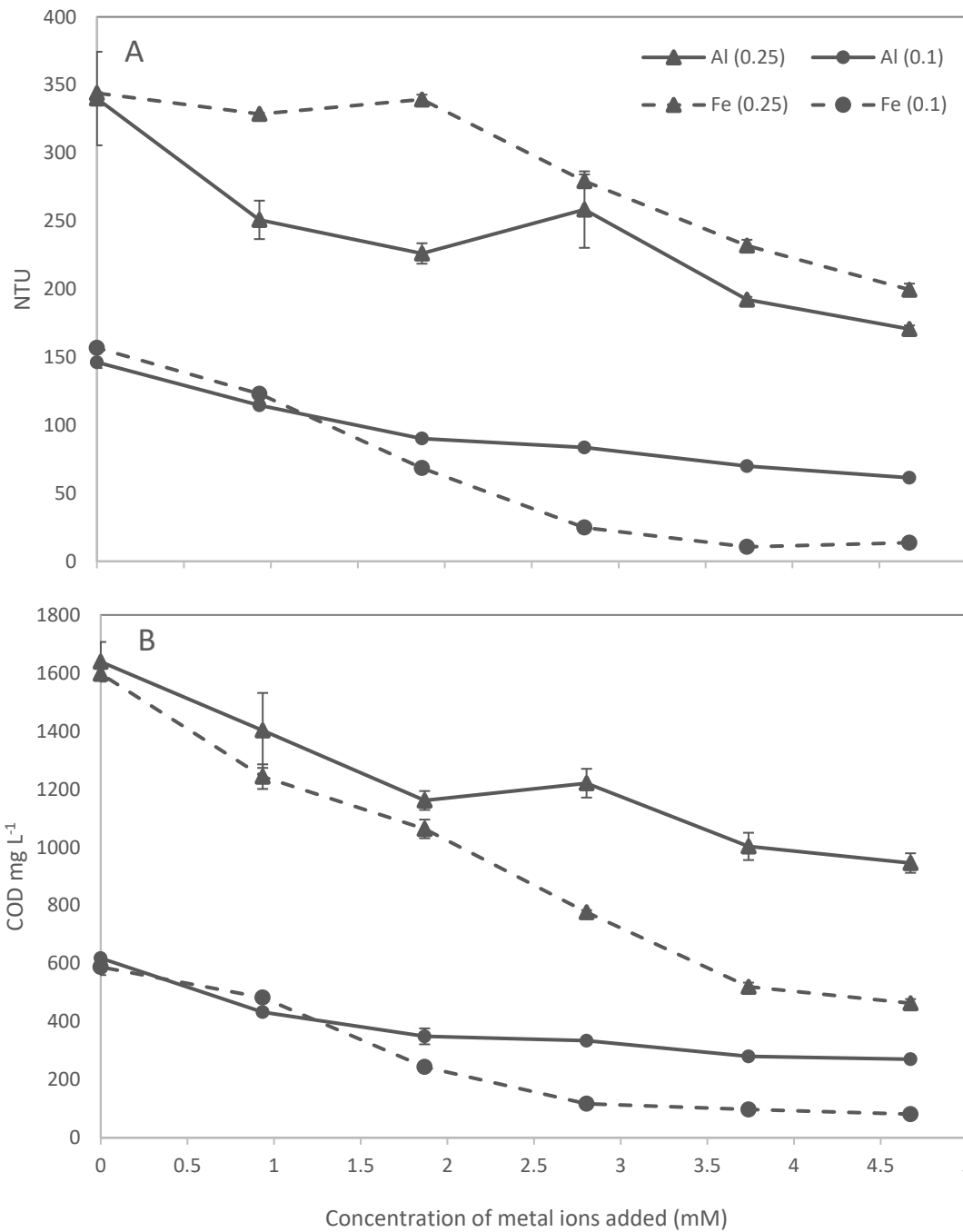
225 **3. Results and discussion**

226 **3.1 Screening tests: Comparisons between Al and Fe electrodes**

227 The overall efficiency of using EC to process wastewaters is highly influenced by the materials
228 used for the construction of the sacrificial anode. In the initial part of this study, we screened
229 the suitability of Fe and Al anodes for EC of food waste digestate due to the relatively low-
230 cost and wide availability of these metals (Gönder et al., 2017; Hakizimana et al., 2017). During
231 this screening we applied the use of a handheld Aerobie filter press as an inexpensive solution
232 for dewatering small volumes of CDEC treated samples. Figure 3 depicts the influence of CDEC
233 on the reduction of turbidity and COD in the filtrate from the handheld filter press. CDEC was
234 able to remove turbidity for all combinations of digestate dilutions and anode materials
235 tested. The nephelometric turbidity units (NTU) in the recovered filtrate was reduced by up
236 to 49.8% and 50.0% after treating the 0.25 and 0.1 digestate dilutions with 4.66 mM Al,
237 respectively. A CDEC Fe coagulant concentration of 4.66 mM reduced the NTU measured in
238 in 0.25 and 0.1 digestate dilutions by up to 41.9% and 91.3%, respectively. The highest %

239 reductions of NTU are similar to those observed in a previous study by Mores et al., (2016) in
240 which Fe anodes were used to remove 91% of turbidity from swine manure digestate from a
241 UASB. During treatment of the 0.25 diluted samples, NTU did not always decrease after higher
242 concentrations of metal ions were added by CDEC (Figure 3a). When adding between 0 and
243 1.86 mM Fe to the 0.25 diluted samples there was no substantial decrease in the mean NTU
244 (Figure 3a). Furthermore, the mean NTU in the filtrate increased from 226 to 258 between
245 1.86 mM and 2.80 mM dosages of Al. One explanation for these non-linear relationships
246 between CDEC metal ion concentration and NTU removal is a shift between two zones in the
247 sequence of increasing coagulant concentration outlined by (Bukhari, 2008) as (1) insufficient
248 dosing, (2) destabilisation, (3) re-stabilisation and (4) sweep-floc coagulation.

249 The concentration of COD in filtrate was reduced by Fe and Al during CDEC experiments
250 (Figure 3b). After applying the higher doses of metal cations, Fe outperformed Al in terms of
251 COD removal. The addition of 4.66 mM of Al by CDEC reduced the concentration of COD in
252 the filtrate by 693 mg L⁻¹ and 348 mg L⁻¹ in 0.25 and 0.1 diluted digestate, respectively. In
253 comparison, the equivalent concentrations of Fe from CDEC reduced the concentration of
254 COD by 1134 mg L⁻¹ and 506 mg L⁻¹, respectively.



255

256

257 **Figure 3. Total nephelometric turbidity units (NTU) (A) and chemical oxygen demand (COD)**
 258 **(B) in filtrate from 0.25 and 0.1 dilutions of digestate after treatment with Al and Fe**
 259 **coagulant-dosing electrocoagulation (CDEC). Data points represent means of triplicate**
 260 **independent repeats ± standard deviation (error bars). Where error bars are not seen, they**
 261 **were smaller than the symbols used to represent the mean values.**

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267 **3.2 Continuous-flow electrocoagulation and filter-press dewatering**

268 Preliminary experimental trials to treat undiluted digestate by using CFEC were ineffective
269 and inoperable at bench-scale due to the rapid fouling of Al and Fe anodes. This fouling caused
270 physical blockages to the throughput of digestate in the EC chamber. Therefore, CFEC was
271 operated using digestate diluted to concentrations of 0.1 and 0.25 as per previous CDEC
272 experiments.

273 During CDEC experiments it was determined that Fe anodes were more efficient than Al at
274 removing COD from the digestate. Furthermore, Fe is less expensive than Al and therefore Fe
275 was selected for the CFEC trials (Gatsios et al., 2015).

276 **3.2.1 Soluble COD and turbidity of filtrate**

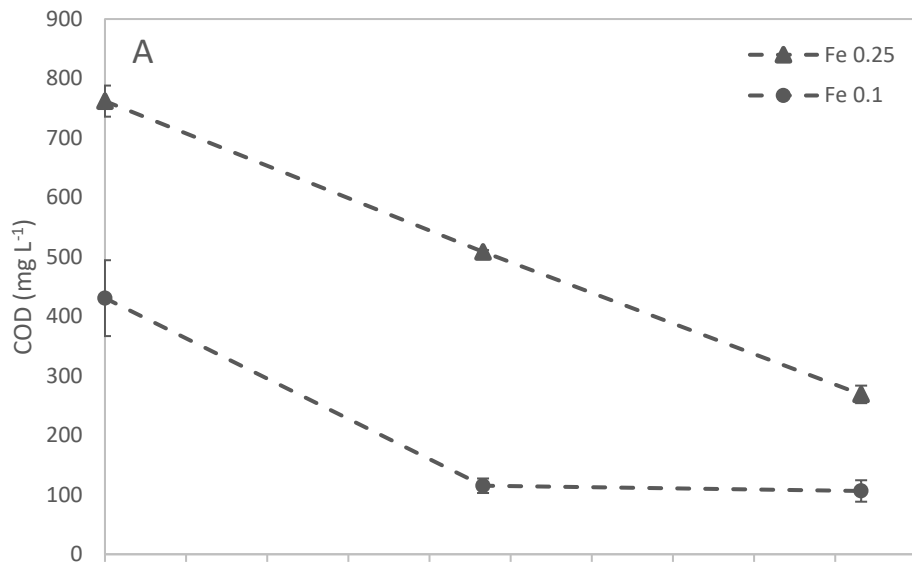
277 When processing digestate with CFEC, increasing the concentration of Fe addition decreased
278 the concentration of COD in the effluent collected from the filter press (Figure 4a). After
279 dewatering, the filtrate from the 0.25 and 0.1 digestate control samples contained 762 mg L⁻¹
280 and 430 mg L⁻¹ of COD, respectively. The filtrate from the dewatered 2.33 mM Fe CFEC
281 treated 0.25 or 0.1 dilutions contained 508 mg L⁻¹ and 115 mg L⁻¹ of COD, respectively.
282 Furthermore, the application of 4.66 mM Fe reduced the concentration to 106 mg L⁻¹ of COD
283 in the 0.1 diluted digestate and 268 mg L⁻¹ of COD in the 0.25 diluted digestate samples.
284 Therefore, the highest removal of COD per mM of Fe added was 135 mg L⁻¹ and this was
285 achieved when treating the 0.1 dilution with 2.33 mM of Fe. The highest absolute COD
286 removal was 64.8% which occurred after applying 4.66 mM Fe to treat the 0.25 diluted
287 digestate. The % of COD removal observed in this study is comparable to other investigation
288 in which EC has been used to treat effluents from olive mills (Tezcan Ün et al., 2006), pistachio
289 processing plants (Ozay et al., 2018), landfill sites (Tezcan Un et al., 2018), municipal

290 wastewater plants (Devlin et al., 2019) and potato chip manufacturing waste (Koby et al.,
291 2006).

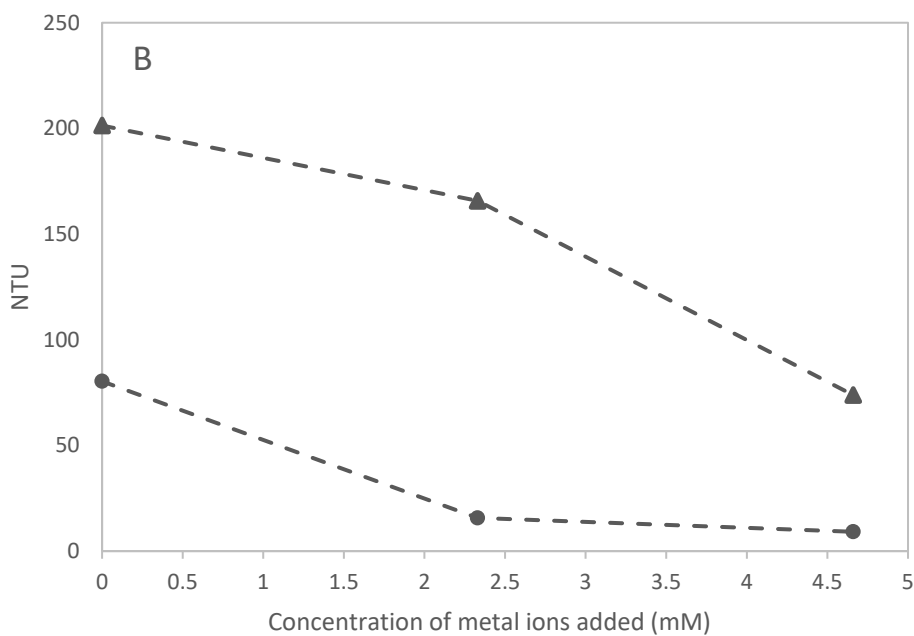
292 Above 2.33 mM Fe treatment, there was no substantial decrease in the concentration of COD
293 in the 0.1 diluted digestate. These findings suggest that 2.33 mM Fe was sufficient to
294 coagulate the concentration of COD-containing compounds in the 0.1 dilution that are able
295 to bind with Fe. Therefore, the results from treatment of the 0.1 dilution of digestate lead us
296 to hypothesise that, beyond the technological hurdle of plate fouling at bench-scale, 1.3 g of
297 Fe would be sufficient to remove all soluble Fe-reactive COD-containing compounds found in
298 1 L of undiluted digestate. Residual COD found in the 2.33 mM and 4.66 mM Fe CFEC treated
299 0.1 diluted samples can be attributed to organic compounds (e.g. glucose, lactose, sucrose)
300 which are known to occur in digestate but do not react or have limited reactivity with Fe (II)
301 or Fe (III) (Moreno-Casillas et al., 2007).

302 A similar trend to COD removal was also observed for the removal of turbidity by Fe CFEC
303 (Figure 3b). NTU in the filtrate effluent from dewatering was reduced by as much as 63.9%
304 and 75.3% in comparison to the 0.25 and 0.1 digestate controls, respectively. The inverse
305 trend between Fe addition and COD concentration in the filtrate is consistent with
306 observations made during the CDEC screening tests. The differences in absolute
307 concentrations of COD between 4.66 mM Fe CDEC and CFEC tests can be attributed to the
308 change in the filter paper type from Aeropress® to Fisher Brand QT280 which had the
309 necessary diameter for use in the dewatering filter-press (Figure 2).

310



311



312

313 **Figure 4. Total chemical oxygen demand (COD) (A) and nephelometric turbidity units (NTU)**
 314 **(B) in filtrate from 0.25 and 0.1 dilutions of digestate after treatment with Fe continuous-**
 315 **flow electrocoagulation (CFEC). Data points represent means of triplicate independent**
 316 **repeats ± standard deviation (error bars). Where error bars are not seen, they are smaller**
 317 **than the symbols used to represent the mean values.**

318

319 **3.2.3 Quantification of solids and filtrate recovery**

320 The application of CFEC reduced the concentrations of TSS and TS in the filtrate recovered
 321 from both 0.25 and 0.1 dilutions of digestate. CFEC using 4.66 mM of Fe reduced the TS in the
 322 filtrate by up to 33.3% and 19.5% in the 0.25 and 0.1 dilutions of digestate, respectively. This

323 reduction of TS in the filtrate was associated with a rise in the mean average TS recovered in
 324 the pressed cake mass. The TS recovered in the filter pressed cake increased stepwise with
 325 the concentration of Fe applied during CFEC (Tables 2 and 3). The relationship between Fe
 326 addition and TSS recovery from the liquid fraction during filtration can be explained by the
 327 growth of particles caused by enhanced coagulation of solids when higher concentrations of
 328 Fe cations were added.

329 The majority of previously reported EC studies have applied batch operation to wastewaters.
 330 Here we applied CFEC. However, fouling of anodes over time (caused by the binding of solids
 331 from wastewater to the metal plates) is known to restrict electrode dissolution during CFEC
 332 treatment of high-strength effluents (Schulz et al., 2009). To alleviate the bench-scale
 333 challenges of EC associated with plate-fouling the digestate was diluted prior to CFEC
 334 treatment. Even though the dilution of the digestate requires an initial addition of water into
 335 the process, the amount of total filtrate recovered from the solids dewatering filter-press
 336 exceeded the volume of water used for dilution (Tables 2 and 3). Although it was beyond the
 337 scope of this proof of concept study, further investigation is now required to test and optimise
 338 the potential for recirculation of the filtrate within the treatment process for dilution of
 339 incoming digestate.

Table 2. Solids and liquids recovery following continuous flow electrocoagulation and dewatering of 0.25 diluted digestate. Individual values represent means of triplicate independent repeats \pm standard deviation.

Fe treatment [mM]	Total Solids in filtrate (g L ⁻¹)	Total Volatile Solids in filtrate (g L ⁻¹)	Total Suspended Solids in filtrate (g L ⁻¹)	Total Solids in filter pressed cake (g L ⁻¹ of digestate treated)	Total filtrate Recovery (ml L ⁻¹ of digestate treated)
(0) Control	1.80 \pm 0.26	1.27 \pm 0.10	0.165 \pm 0.01	2.8 \pm 0.1	923 \pm 8
2.33	1.43 \pm 0.09	0.90 \pm 0.13	0.104 \pm 0.01	2.9 \pm 0.1	963 \pm 29
4.66	1.21 \pm 0.07	0.68 \pm 0.08	0.077 \pm 0.01	3.2 \pm 0.2	936 \pm 24

Table 3. Solids and liquids recovery following continuous flow electrocoagulation and dewatering of 0.1 diluted digestate. Individual values represent means of triplicate independent repeats \pm standard deviation.

Fe treatment [mM]	Total Solids in filtrate (g L ⁻¹)	Total Volatile Solids in filtrate (g L ⁻¹)	Total Suspended Solids in filtrate (g L ⁻¹)	Total Solids in filter pressed cake (g L ⁻¹ of digestate treated)	Total filtrate Recovery (ml L ⁻¹ of digestate treated)
0 (Control)	1.28 \pm 0.02	0.77 \pm 0.03	0.181 \pm 0.00	1.0 \pm 0.1	953 \pm 41
2.33	1.06 \pm 0.04	0.52 \pm 0.03	0.108 \pm 0.01	1.1 \pm 0.2	986 \pm 11
4.66	1.03 \pm 0.06	0.58 \pm 0.08	0.068 \pm 0.00	1.4 \pm 0.1	962 \pm 3

340

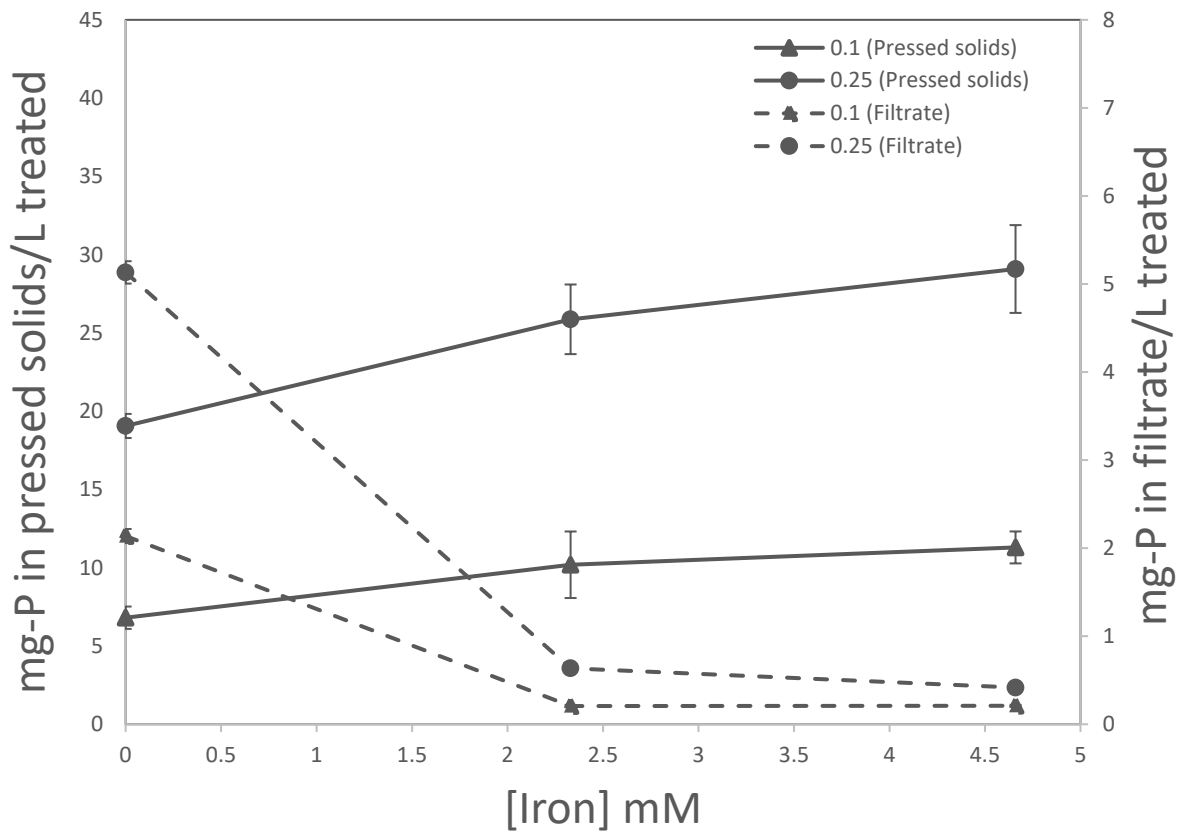
341 3.2.4 Composition of cake and filtrate

342 3.2.4.1 Recovery of phosphorous and nitrogen

343 CFEC successfully increased both the total yield and fraction of P recovered in filter-pressed
 344 cake. The average increase in the total yield of P recovered in cake solids, collected per litre
 345 of digestate treated, was 1.65 and 1.52 fold compared to the controls for the 0.1 and 0.25
 346 dilutions, respectively (Figure 5). Furthermore, the application of 4.66 mM Fe by CFEC
 347 increased the proportion of total P in the recovered solids from 6.8 ± 0.0 mg kg-TS⁻¹ in the
 348 controls to 8.0 ± 0.1 mg kg-TS⁻¹ and 9.0 ± 0.2 mg kg-TS⁻¹ when treating the 0.1 and 0.25
 349 dilutions of digestate, respectively. Soluble P in the filtrate decreased by up to 92% and 90%
 350 for the 0.25 and 0.1 dilutions, respectively. A decrease of total P in the corresponding filtrates
 351 confirmed that CFEC caused the precipitation of soluble P-containing compounds and enabled
 352 their recovery by filtration (Figure 5). The most efficient recovery of P per Fe added was 26.42
 353 g Fe g-P⁻¹ recovered and this was achieved when 2.33 mM Fe was applied to the 0.25 dilution
 354 of digestate.

355 In all CFEC treatments and controls the pH of the filtrate effluent remained between 7.19 and
 356 7.73. These pH values are firmly within the optimum operational pH ranges of 5-9 for Fe EC

357 (Huang et al., 2017; Moussa et al., 2017). Due to the pH of the samples being above neutral
 358 throughout CFEC treatment and settlement, precipitation of phosphate by Fe^{2+} to produce
 359 $\text{Fe}_3(\text{PO}_4)_2$ was likely the primary mechanism of P removal from the soluble phase (Omwene et
 360 al., 2018). Soluble P removal of c.90% from food waste digestate (in the current study) is
 361 comparable to previous EC studies on laundry wastewater (Janpoor et al., 2011), dairy
 362 wastewater (Kuokkanen et al., 2015) and dairy manure (Zhang et al., 2016) in which removal
 363 of P from the soluble phase ranged between 90.9-96.7%.



364
 365 **Figure 5. Total P in filtrate and cake solids recovered from 0.1 and 0.25 dilutions of digestate**
 366 **after treatment with Fe continuous-flow electrocoagulation (CFEC). Data points represent**
 367 **means of triplicate independent repeats \pm standard deviation (error bars). Where error bars**
 368 **are not seen, they are smaller than the symbols used to represent the mean values.**
 369
 370

371 The proportion of total N in the recovered pressed cake solids ranged between 7.34% and
372 8.94% (d.w) for all samples and therefore was similar between CFEC treatments and controls.
373 Furthermore, NH_4^+ concentration in the digestate filtrate was unaffected by EC treatment.
374 Previously reported electrochemical treatment has been shown to convert NH_4^+ to NO_3^- and
375 N_2 gas via oxidation (Ghimire et al., 2019; Li and Liu, 2009). However, in a previous
376 investigation by Liu and Liu (2016) NH_4^+ removal also remained low while using EC to treat
377 anaerobic digestate. Here, EC was not expected to convert NH_4^+ to nitrogen gas, as
378 parameters were not optimised to generate the hypochlorite required to drive the reaction
379 (Ding et al., 2021).

380 This NH_4^+ -containing filtrate has had the majority of soluble P removed which opens the
381 possibility of differentiation between the two nutrients for agricultural purposes.
382 Furthermore, the N could be removed from the filtrate in an additional processing stage using
383 conventional N-removal techniques, such as physicochemical, biological, or electrochemical
384 treatments (Mook et al., 2012).

385 **3.2.4.2 Recovery of metals**

386 Digestate can contain potentially toxic elements (PTE) (e.g. Cr, Mo, Pb) which if in high
387 concentrations could be hazardous to animals and plants (DEFRA, 2018). Hence, the amount
388 of digestate which is applied to soils must be controlled to ensure that spreading of PTE isn't
389 environmentally hazardous. Therefore, as part of this study, we used ICP-MS to measure the
390 concentrations of a range of metals in the cake and soluble phases, following EC treatment.
391 Fe CFEC was found to cause negligible increases to the concentrations of PTE Cr and As in
392 pressed cake solids of up to $71.1 \pm 9.1 \text{ mg kg}^{-1}$ and $0.6 \pm 0.3 \text{ mg kg}^{-1}$, respectively. The mean
393 average concentrations of Cu, Ni and Zn were marginally increased after Fe CFEC by up to 4.1

394 $\pm 12.0 \text{ mg kg}^{-1}$, $26.3 \pm 2.8 \text{ mg kg}^{-1}$ and $10.8 \pm 20.5 \text{ mg kg}^{-1}$, respectively. Following Fe CFEC
395 treatment, no increases in the concentrations of PTE Cd, Mo or Pb in cake were detected.

396 N application in UK agriculture is limited to a maximum of 250 kg N per hectare per year and
397 therefore this value was used to calculate the maximum amounts of each digestate cake solids
398 (prepared by CFEC and dewatering) that could be applied to UK farming land (DEFRA, 2015).
399 When using any sets of EC treatments tested in this study, PTE Cu, Ni, Zn, As, Cd, Cr, Mo and
400 Pb would all remain below permitted application limits in a scenario in which recovered
401 pressed cake digestate is applied at the maximum allowed rate of 250 kg N per hectare per
402 year.

403 Although Fe is not listed by the UK government as a PTE, it is notable that all the CFEC
404 treatments caused a substantial increase of Fe in pressed cake solids, compared to the control
405 which contained $0.08 \pm 0.00\%$. The pressed cake solids collected from the 0.1 diluted
406 digestate samples contained $10.57 \pm 0.22\%$ and $17.95 \pm 0.44\%$ Fe after applying 2.33 mM and
407 4.66 mM Fe CFEC treatment, respectively. A similar trend was observed for the fractions of
408 Fe in the pressed cake solids derived from the 0.25 diluted samples which contained $3.7 \pm$
409 0.32% and $8.12 \pm 0.40\%$ following 2.33 mM and 4.66 mM Fe CFEC treatment, respectively.
410 These increases in Fe within the filter pressed cake solids can be attributed to the use of steel
411 anodes during CFEC. However, although CFEC increased the Fe concentration in the filter
412 pressed cake solids it should be noted that Fe is highly abundant in soils globally (Towett et
413 al., 2015). Therefore, in many locations, the relatively small addition of Fe into pressed cake
414 solids (caused by CFEC) is unlikely to have negative implications when using the cake as an
415 agricultural fertiliser. Furthermore, the application of Fe to land has been reported to be
416 advantageous for agricultural purposes (Brown et al., 2012). In all CFEC treatments the

417 residual concentration of Fe in the recovered filtrate remained below the typical median Fe
418 concentration of 0.7 mg L⁻¹ found in river water (WHO, 1996).

419 **4. Estimation of Fe addition costs**

420 The operational cost of CFEC is associated with electrical power consumption and anode
421 materials. Although EC technology continues to evolve and become more efficient, at the time
422 of writing, the cost for the Fe dosing concentrations used in this study are estimated to be
423 \$8.6x10⁻⁴ US g-Fe⁻¹ and \$1.2x10⁻³ US g-Fe⁻¹ for 2.33 mM and 4.66 mM, respectively. Therefore,
424 it is estimated that treating 10 m³ of 0.1 or 0.25 diluted digestate with 2.33 mM or 4.66mM
425 of Fe would cost \$1.13 US and \$3.17 US, respectively. These estimations are based on an
426 average cost of \$0.19 US kWh⁻¹ and values have been provided based on a pilot-scale Elentec
427 Ltd. system in operation at the time of writing.

428 **5. Conclusions**

429 In this study we demonstrated the potential of EC for the recovery of nutrients and other
430 solids from food waste digestate. Fe was found to outperform Al in terms of COD removal
431 from the soluble phase of food waste digestate. CFEC using 2.33 mM Fe and a 0.25 dilution of
432 digestate was the optimal treatment tested. This treatment achieved the highest P recovery
433 from the soluble phase of 22.4 mg-P per g-Fe added, the greatest reduction of solids in the
434 filtrate (2.84 g per g-Fe added), substantial COD removal (of 254 mg L⁻¹) and was estimated to
435 be the least expensive ratio of Fe addition to digestate.

436 Our results indicate that, in a scenario whereby recovered cake from EC-treated food waste
437 digestate is applied to soil at a rate of 250 kg N per hectare per year, Cu, Ni, Zn, As, Cd, Cr, Mo
438 and Pb would remain below their permitted application limits in the UK. Plant-growth trials

439 are now required in further work to examine the quality of the nutrient-enhanced pressed
440 cake solids from food waste digestion as a soil fertiliser for agricultural purposes.

441 **Acknowledgments**

442 The authors would like to acknowledge research grant funding provided by the EPSRC for the
443 *Redivivus* project (grant code: EP/M007359/1). We would also like to thank Elentec Ltd. for
444 contributing staff and equipment, and Michael A Cooper for his assistance.

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