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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

1. Digestates from anaerobic digestion (AD) of food waste contain fertiliser nutrients (such as
2. P~~phosphorous~~ and N~~nitrogen~~) which are valuable for agricultural purposes and can be
3. environmentally hazardous if disposal is uncontrolled. Here, we applied electrocoagulation
4. (EC) for treatment of digestates, to separate liquids and nutrient-rich solids. Coagulant-dosing
5. electrocoagulation (CDEC) was used to compare Al and steel anodes for treatment of
6. digestate from AD fermenters fed a controlled diet representative of food waste. When
7. applying metal dosing concentrations of 0 - 4.66 mM, Fe was found to be up to 29.8% superior
8. to Al in terms of aiding removal of chemical oxygen demand (COD). To mitigate plate fouling,

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

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

# 1. Introduction

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

Although food waste digestate is rich in valuable fertiliser nutrients, its application in agriculture remains challenging. The nutrients in raw digestates are contained in large amounts of water. The overall volume of water not only has negative implications for the costs associated with digestate transportation but also increases the possibility of environmental damage via eutrophication or contamination of natural water systems (Tampio et al., 2016). To enable targeted fertiliser application and limit the environmental risk, suitable digestate treatment methods are necessary to extract fertiliser nutrients (such as N, P and K) out of solution. Typically, solid and liquid separation methodologies initially use a solids thickening technology (which often consists of settling, filtration or centrifugation) and incorporates the use of a polyelectrolyte or metal chloride coagulants (e.g. ferric chloride) (Monfet et al., 2018). The thickening of solids in digestate is usually followed by a dewatering step, which is commonly carried out by using either a filter press or centrifugation (Knocke et al., 1993; Wakeman, 2007).

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

At the cathode:

2H2O(l) + 2e- → H2(g) + 2OH-(aq) (1)

At the anode:

Al(s) → Al3+(aq) +3e-  (2) Fe(s) → Fe2+(aq) + 2e-  (3)

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

*w = (I t M)/(z F)*  (4)

Where *I* is the current (A), *t* is the process time (s), *M* is the molar mass of the electrode metal (g mol-1), *z* is the valence of the anode metal and *F* is Faraday’s constant (96,485 C mol-1).

Once in the digestate, the metal ions react with OH- groups to form hydroxides which can coagulate solids by complexation or electrostatic attraction (Hakizimana et al., 2017). Electrodes are most often constructed out of Fe and Al because these metals have high valence, are cost effective and widely available (Hakizimana et al., 2017; Tirado et al., 2018). Furthermore, the respective hydroxides (e.g. Al(OH)3 and Fe(OH)2 are highly insoluble (Ksp 3x10-34 and 8x10-16, respectively). Multiple factors influence the cost and performance of EC treatment such as the metal ion concentration (Mameri et al., 1998), initial pH (Tezcan Un et al., 2018), retention time (Tezcan Un et al., 2018), electrode material (Devlin et al., 2019), EC chamber design (Kabdaşlı et al., 2012) and salt concentration (Yıldız et al., 2008).

EC technology can be operated in various treatment configurations which include ‘coagulant dosing’ (where the electrodes are used to generate a concentrated solution of coagulant which is then introduced into the effluent stream), ‘batch’ (where the electrodes are corroded directly into the effluent but treatment is carried out by processing discrete volumes of effluent at a time) (Tezcan Ün et al., 2006; Yıldız et al., 2008) and ‘continuous flow’ (where electrodes are operating continuously and in direct contact with the effluent as it flows through the reactor/cell) (Benazzi et al., 2015; Makwana and Ahammed, 2016). Each configuration has specific advantages. Coagulant dosing limits direct contact of digestate with plates and mitigates electrode fouling. Batch treatment is carried out by submersing the anode and cathode into the digestate and requires minimal equipment. Continuous flow EC enables uninterrupted processing of large treatment loads (Benazzi et al., 2015). Previously, EC has been successfully applied to treat a range effluent streams including those from dairies and slaughterhouses (Reilly et al., 2019), the textile industry (Alinsafi et al., 2005), landfill leachates (Li et al., 2011) and wastewater (Kuokkanen et al., 2013). EC is potentially advantageous over conventional coagulation methods for digestate treatment as it excludes the addition of synthetic polymers to digestate, which minimises the potential for contamination of land during spreading of the recovered solids onto agricultural soils (Lapointe and Barbeau, 2019). Furthermore, metal electrodes occupy less space than their corresponding metal chloride coagulants. The reduced footprint is beneficial for both transportation and storage requirements. Additionally, storage of electrodes is significantly safer than that of large volumes of metal chloride solutions.

A limited number of studies have successfully investigated the use of EC to treat digestates from AD. To date, these studies have mainly focused on effluents from AD plants processing sewage and municipal solid waste (MSW) (Fernandes et al., 2017; Huang et al., 2017; Makwana and Ahammed, 2016; Olvera-Vargas et al., 2019). For example Huang et al. (2017) demonstrated that the use of Fe2+ EC to recover phosphate from anaerobic sludge worked optimally between pH 6-8 and concluded that the recovery of phosphate by EC using electrodes made of Fe was more efficient than using those made of Al. Makwana and Ahammed (2017) and Fernandes et al., (2017) reported that chemical oxygen demand (COD) removal of 71% and 80% could be achieved from digestates derived from urban wastewater and MSW, respectively. Makwana and Ahammed (2016) described a continuous process which they applied to effluent from an upflow anaerobic sludge blanket (UASB) reactor treating municipal wastewater. When using a current density of 20 mA cm-2 the process removed 67.2% COD and reduced initial phosphate concentrations (ranging between 4.5-6.4 mg L-1) down to 0.57 mg L-1.

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

# 2. Materials & Methods

## 2.1 Digestate preparation and characteristics

The digestate used in this study was sourced from laboratory-scale anaerobic digesters, which were operated in duplicate. The anaerobic digesters were inoculated with sludge digestate sourced from a mesophilic digester operating at 35°C on Monkmoor sewage treatment works (Shrewsbury, UK). Each laboratory-scale digester was fed a synthetic recipe representing food waste as per Jobling-Purser (2015). Feedstock was prepared weekly and stored at 5°C. Each laboratory anaerobic digester was 26L and had a working volume of 23L. The digesters were fed daily at a rate of 2.5 Kg-VS m-3 d -1 of synthetic food waste and using a hydraulic retention time (HRT) of 20 days. The digesters were mesophilic (35°C) and stirred at 100 rpm; they were operated for 3 HRT to ensure steady-state operation before digestate was used in EC trials.

Digestate was collected each day, pooled and refrigerated (at 5°C) for no more than 7 days prior before being passed through a 500 μm sieve and used in EC trials. The characteristics of 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-CaCO3 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 |
| NH4-N (soluble) | mg L-1 | 383 | ± | 4 |
| NO3-N (soluble) | mg L-1 | 20.6 | ± | 3.1 |

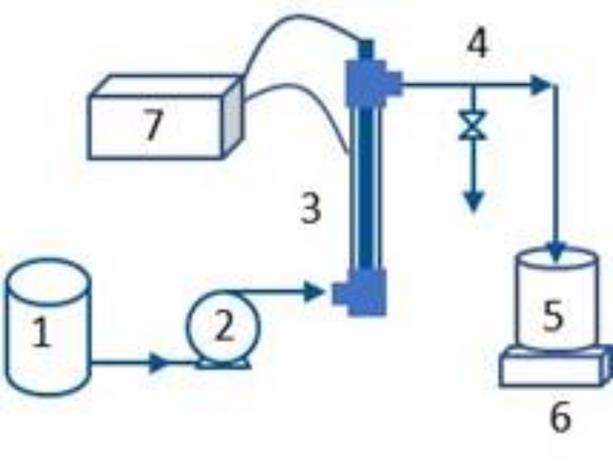
## 2.2 Electrocoagulation processing

### 2.2.1 Electrocoagulation equipment

Electrochemical experiments were performed using a benchtop EC system (Elentec Ltd., UK). The system included a circulation pump (Model 630S, Watson Marlow Ltd., UK) and power supply (EA Elektro-Automatik EA-PS 2042-10B, RS Components Ltd., UK). A simplified flow diagram is presented in Figure 1. The EC cell consisted of a tubular cathode in stainless steel

(SS 304, ID 19 mm, length 25.8 cm) fitted with ½” BSP fittings which located the anode along the middle axis of the cell. Anodic material was either mild steel (12mm diam., RS Components Ltd., UK) or aluminium (6082-T6, 12mm diam., RS components). The active surface area of the anode was 97.3 cm2.

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



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

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

Screening experiments were completed to examine the feasibility of EC for the treatment of food waste digestate and to select suitable parameters for subsequent dewatering experiments. CDEC was used to create a coagulant solution by releasing Fe and Al ions from the anode into solution, which was then added to digestate. This coagulant was produced as follows; 17.1 mM NaCl (with a conductivity of 2 mS cm-1) was pumped through the EC cell. At constant flow rate, the current input was varied according to Faraday’s equation (equation 4) to generate concentrations of Fe or Al coagulant solutions. Either 100 ml or 250 ml of digestate were made up to 1L through addition of generated electrolyte solution, resulting in a 0.1 and 0.25 dilution of digestate, respectively. The final coagulant-dosing concentration of Fe and Al in the diluted samples were 0.933, 1.87, 2.80, 3.73, 4.66 mM. Control samples were prepared which contained the volumes of digestate described above but with 17.1 mM NaCl

(instead of coagulant solution). Each CDEC condition was repeated in triplicate. Therefore, CDEC experimentation included a total sample size of 72 (Supplementary Table A).

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

### 2.2.3 Continuous-flow electrocoagulation (CFEC)

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

## 2.3 Dewatering of CFEC samples

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

## 2.4 Analytical Methods

Total solids (TS) and volatile solids (VS) were measured gravimetrically according to the standard method (APHA, 1989). Total suspended solids in samples were quantified according to EPA Method 160.2 using 0.7μm Merck™ AP4004705 filters (Fisher Scientific, UK) (EPA, 1971). Conductivity was measured by using a Hanna HI 86304 conductivity meter equipped with a HI-7632D/1 probe. A Jenway 3510 pH meter was used for pH measurements (Jenway,

UK). Alkalinity was quantified by using a KEM AT-710 auto-titrator (Kyoto Electronics Manufacturing Co. Ltd., Japan) and the titration method of Jenkins et al. (1983). Turbidity was analysed by use of a ThermoScientific Eutech TN-100 meter (Fisher Scientific, UK). COD was determined using a potassium dichromate method Hach Lange LCI 400 assay kit (Hach Lange,

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

# 3. Results and discussion

## 3.1 Screening tests: Comparisons between Al and Fe electrodes

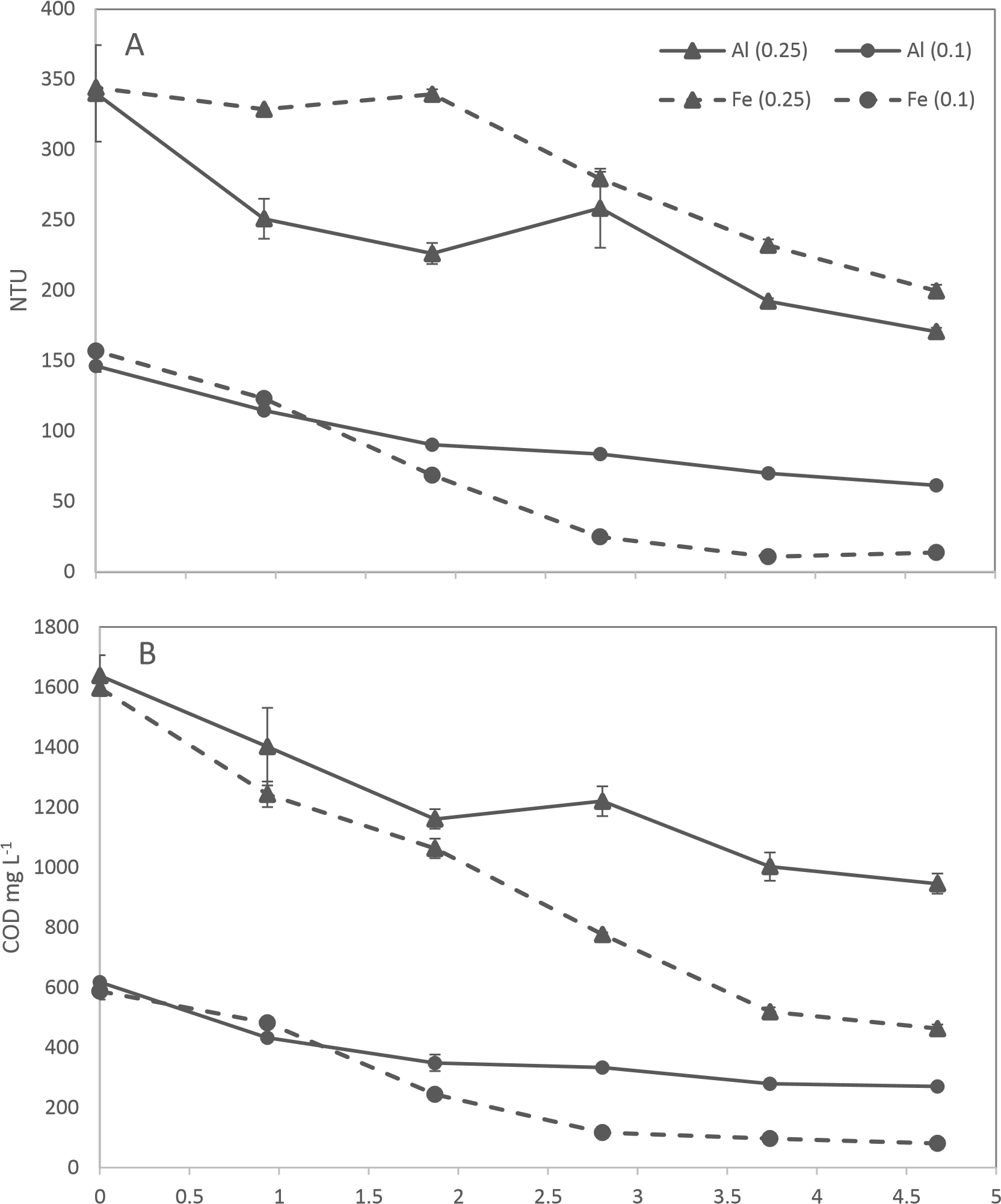
The overall efficiency of using EC to process wastewaters is highly influenced by the materials used for the construction of the sacrificial anode. In the initial part of this study, we screened the suitability of Fe and Al anodes for EC of food waste digestate due to the relatively lowcost and wide availability of these metals (Gönder et al., 2017; Hakizimana et al., 2017). During this screening we applied the use of a handheld Aerobie filter press as an inexpensive solution for dewatering small volumes of CDEC treated samples. Figure 3 depicts the influence of CDEC on the reduction of turbidity and COD in the filtrate from the handheld filter press. CDEC was able to remove turbidity for all combinations of digestate dilutions and anode materials tested. The nephelometric turbidity units (NTU) in the recovered filtrate was reduced by up to 49.8% and 50.0% after treating the 0.25 and 0.1 digestate dilutions with 4.66 mM Al, respectively. A CDEC Fe coagulant concentration of 4.66 mM reduced the NTU measured in in 0.25 and 0.1 digestate dilutions by up to 41.9% and 91.3%, respectively. The highest % reductions of NTU are similar to those observed in a previous study by Mores et al., (2016) in which Fe anodes were used to remove 91% of turbidity from swine manure digestate from a UASB. During treatment of the 0.25 diluted samples, NTU did not always decrease after higher concentrations of metal ions were added by CDEC (Figure 3a). When adding between 0 and

1.86 mM Fe to the 0.25 diluted samples there was no substantial decrease in the mean NTU (Figure 3a). Furthermore, the mean NTU in the filtrate increased from 226 to 258 between 1.86 mM and 2.80 mM dosages of Al. One explanation for these non-linear relationships between CDEC metal ion concentration and NTU removal is a shift between two zones in the sequence of increasing coagulant concentration outlined by (Bukhari, 2008) as (1) insufficient dosing, (2) destabilisation, (3) re-stabilisation and (4) sweep-floc coagulation.

The concentration of COD in filtrate was reduced by Fe and Al during CDEC experiments

(Figure 3b). After applying the higher doses of metal cations, Fe outperformed Al in terms of COD removal. The addition of 4.66 mM of Al by CDEC reduced the concentration of COD in the filtrate by 693 mg L-1 and 348 mg L-1 in 0.25 and 0.1 diluted digestate, respectively. In comparison, the equivalent concentrations of Fe from CDEC reduced the concentration of

COD by 1134 mg L-1 and 506 mg L-1, respectively.



Concentration of metal ions added (mM)

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

## 3.2 Continuous-flow electrocoagulation and filter-press dewatering

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

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

### 3.2.1 Soluble COD and turbidity of filtrate

When processing digestate with CFEC, increasing the concentration of Fe addition decreased the concentration of COD in the effluent collected from the filter press (Figure 4a). After dewatering, the filtrate from the 0.25 and 0.1 digestate control samples contained 762 mg L1 and 430 mg L-1 of COD, respectively. The filtrate from the dewatered 2.33 mM Fe CFEC treated 0.25 or 0.1 dilutions contained 508 mg L-1 and 115 mg L-1 of COD, respectively. Furthermore, the application of 4.66 mM Fe reduced the concentration to 106 mg L-1 of COD in the 0.1 diluted digestate and 268 mg L-1 of COD in the 0.25 diluted digestate samples. Therefore, the highest removal of COD per mM of Fe added was 135 mg L-1 and this was achieved when treating the 0.1 dilution with 2.33 mM of Fe. The highest absolute COD removal was 64.8% which occurred after applying 4.66 mM Fe to treat the 0.25 diluted digestate. The % of COD removal observed in this study is comparable to other investigation in which EC has been used to treat effluents from olive mills (Tezcan Ün et al., 2006), pistachio processing plants (Ozay et al., 2018), landfill sites (Tezcan Un et al., 2018), municipal wastewater plants (Devlin et al., 2019) and potato chip manufacturing waste (Kobya et al., 2006).

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

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

0

100

200

300

400

500

600

700

800

900

COD (mg L

-

1

)

A

Fe 0.25

Fe 0.1

0

50

100

150

200

250

0

0.5

1

1.5

2

2.5

3

3.5

4

4.5

5

NTU

Concentration of metal ions added (mM)

B

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

### 3.2.3 Quantification of solids and filtrate recovery

The application of CFEC reduced the concentrations of TSS and TS in the filtrate recovered from both 0.25 and 0.1 dilutions of digestate. CFEC using 4.66 mM of Fe reduced the TS in the filtrate by up to 33.3% and 19.5% in the 0.25 and 0.1 dilutions of digestate, respectively. This reduction of TS in the filtrate was associated with a rise in the mean average TS recovered in the pressed cake mass. The TS recovered in the filter pressed cake increased stepwise with the concentration of Fe applied during CFEC (Tables 2 and 3). The relationship between Fe addition and TSS recovery from the liquid fraction during filtration can be explained by the growth of particles caused by enhanced coagulation of solids when higher concentrations of Fe cations were added.

The majority of previously reported EC studies have applied batch operation to wastewaters. Here we applied CFEC. However, fouling of anodes over time (caused by the binding of solids from wastewater to the metal plates) is known to restrict electrode dissolution during CFEC treatment of high-strength effluents (Schulz et al., 2009). To alleviate the bench-scale challenges of EC associated with plate-fouling the digestate was diluted prior to CFEC treatment. Even though the dilution of the digestate requires an initial addition of water into the process, the amount of total filtrate recovered from the solids dewatering filter-press exceeded the volume of water used for dilution (Tables 2 and 3). Although it was beyond the scope of this proof of concept study, further investigation is now required to test and optimise the potential for recirculation of the filtrate within the treatment process for dilution of 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 ± 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 ± 0.26 | 1.27 ± 0.10 | 0.165 ± 0.01 | 2.8 ± 0.1 | 923 ± 8 |
| 2.33 | 1.43 ± 0.09 | 0.90 ± 0.13 | 0.104 ± 0.01 | 2.9 ± 0.1 | 963 ± 29 |
| 4.66 | 1.21 ± 0.07 | 0.68 ± 0.08 | 0.077 ± 0.01 | 3.2 ± 0.2 | 936 ± 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 ± 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 ± 0.02 | 0.77 ± 0.03 | 0.181 ± 0.00 | 1.0 ± 0.1 | 953 ± 41 |
| 2.33 | 1.06 ± 0.04 | 0.52 ± 0.03 | 0.108 ± 0.01 | 1.1 ± 0.2 | 986 ± 11 |
| 4.66 | 1.03 ± 0.06 | 0.58 ± 0.08 | 0.068 ± 0.00 | 1.4 ± 0.1 | 962 ± 3 |

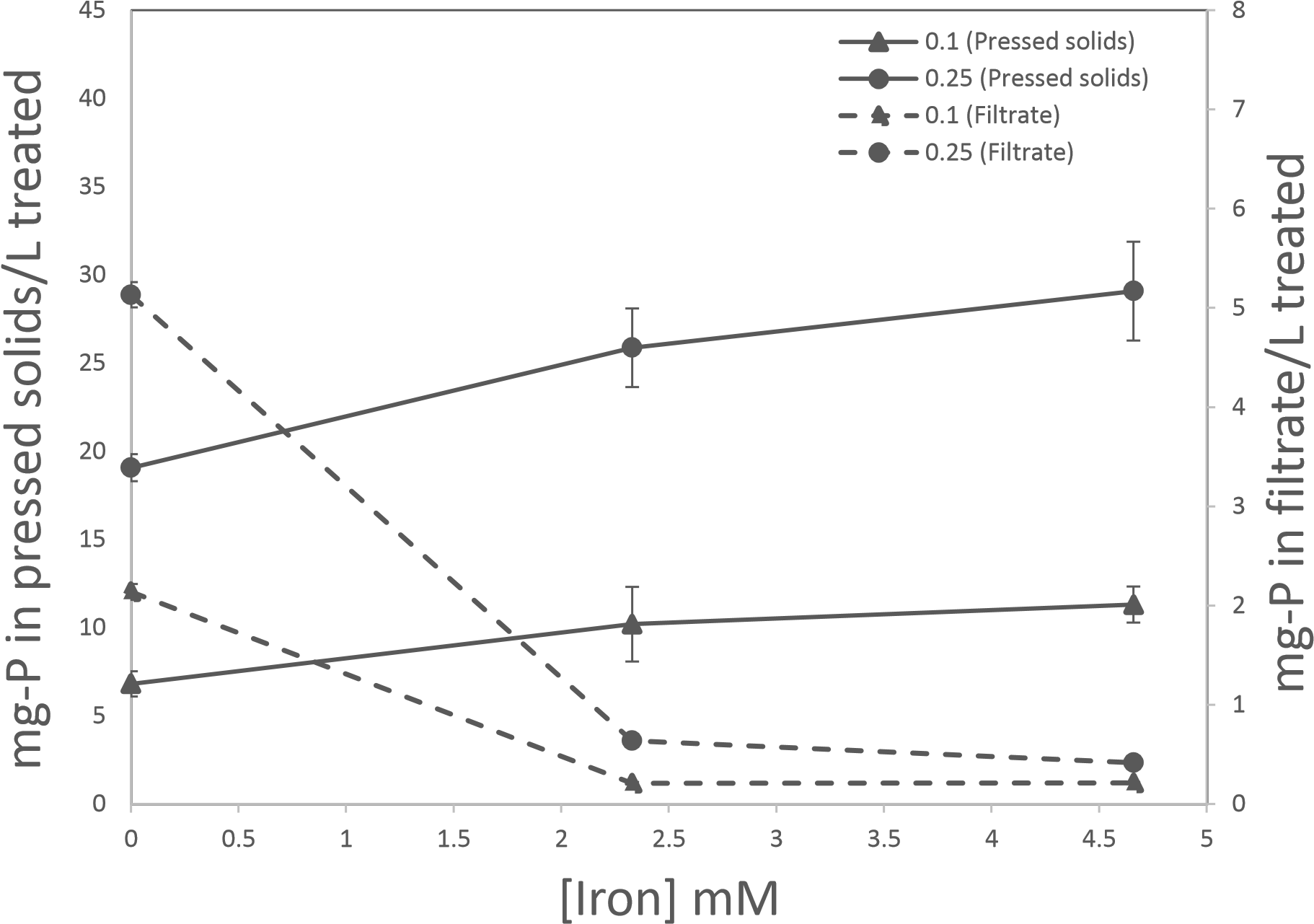
### 3.2.4 Composition of cake and filtrate

#### 3.2.4.1 Recovery of phosphorous and nitrogen

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

In all CFEC treatments and controls the pH of the filtrate effluent remained between 7.19 and

7.73. These pH values are firmly within the optimum operational pH ranges of 5-9 for Fe EC (Huang et al., 2017; Moussa et al., 2017). Due to the pH of the samples being above neutral throughout CFEC treatment and settlement, precipitation of phosphate by Fe2+ to produce Fe3(PO4)2 was likely the primary mechanism of P removal from the soluble phase (Omwene et al., 2018). Soluble P removal of c.90% from food waste digestate (in the current study) is comparable to previous EC studies on laundry wastewater (Janpoor et al., 2011), dairy wastewater (Kuokkanen et al., 2015) and dairy manure (Zhang et al., 2016) in which removal of P from the soluble phase ranged between 90.9-96.7%.



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

The proportion of total N in the recovered pressed cake solids ranged between 7.34% and

8.94% (d.w) for all samples and therefore was similar between CFEC treatments and controls. Furthermore, NH4+ concentration in the digestate filtrate was unaffected by EC treatment.

Previously reported electrochemical treatment has been shown to convert NH4+ to NO3- and N2 gas via oxidation (Ghimire et al., 2019; Li and Liu, 2009). However, in a previous investigation by Liu and Liu (2016) NH4+ removal also remained low while using EC to treat anaerobic digestate. Here, EC was not expected to convert NH4+ to nitrogen gas, as parameters were not optimised to generate the hypochlorite required to drive the reaction (Ding et al., 2021).

This NH4+-containing filtrate has had the majority of soluble P removed which opens the possibility of differentiation between the two nutrients for agricultural purposes. Furthermore, the N could be removed from the filtrate in an additional processing stage using conventional N-removal techniques, such as physicochemical, biological, or electrochemical treatments (Mook et al., 2012).

#### 3.2.4.2 Recovery of metals

Digestate can contain potentially toxic elements (PTE) (e.g. Cr, Mo, Pb) which if in high concentrations could be hazardous to animals and plants (DEFRA, 2018). Hence, the amount of digestate which is applied to soils must be controlled to ensure that spreading of PTE isn’t environmentally hazardous. Therefore, as part of this study, we used ICP-MS to measure the concentrations of a range of metals in the cake and soluble phases, following EC treatment. Fe CFEC was found to cause negligible increases to the concentrations of PTE Cr and As in pressed cake solids of up to 71.1 ± 9.1 mg kg-1 and 0.6 ± 0.3 mg kg-1, respectively. The mean average concentrations of Cu, Ni and Zn were marginally increased after Fe CFEC by up to 4.1 ± 12.0 mg kg-1, 26.3 ± 2.8 mg kg-1 and 10.8 ± 20.5 mg kg-1, respectively. Following Fe CFEC treatment, no increases in the concentrations of PTE Cd, Mo or Pb in cake were detected.

N application in UK agriculture is limited to a maximum of 250 kg N per hectare per year and therefore this value was used to calculate the maximum amounts of each digestate cake solids (prepared by CFEC and dewatering) that could be applied to UK farming land (DEFRA, 2015).

When using any sets of EC treatments tested in this study, PTE Cu, Ni, Zn, As, Cd, Cr, Mo and Pb would all remain below permitted application limits in a scenario in which recovered pressed cake digestate is applied at the maximum allowed rate of 250 kg N per hectare per year. While the annual returns of heavy metals to land are clearly within legislative limits, it must be acknowledged that eventually, year-on-year accumulation could pose a problem. However, this is an issue with many circular agriculture opportunities and appropriate strategies, including strategic application rosters and soil nutrient monitoring, will need to be developed alongside fertiliser nutrient circularity in order to mitigate the problem.

Although Fe is not listed by the UK government as a PTE, it is notable that all the CFEC treatments caused a substantial increase of Fe in pressed cake solids, compared to the control which contained 0.08 ± 0.00%. The pressed cake solids collected from the 0.1 diluted digestate samples contained 10.57 ± 0.22% and 17.95 ± 0.44% Fe after applying 2.33 mM and

4.66 mM Fe CFEC treatment, respectively. A similar trend was observed for the fractions of Fe in the pressed cake solids derived from the 0.25 diluted samples which contained 3.7 ±

0.32% and 8.12 ± 0.40% following 2.33 mM and 4.66 mM Fe CFEC treatment, respectively. These increases in Fe within the filter pressed cake solids can be attributed to the use of steel anodes during CFEC. However, although CFEC increased the Fe concentration in the filter pressed cake solids it should be noted that Fe is highly abundant in soils globally (Towett et al., 2015). Therefore, in many locations, the relatively small addition of Fe into pressed cake solids (caused by CFEC) is unlikely to have negative implications when using the cake as an agricultural fertiliser. Furthermore, the application of Fe to land has been reported to be advantageous for agricultural purposes (Brown et al., 2012). In all CFEC treatments the residual concentration of Fe in the recovered filtrate remained below the typical median Fe concentration of 0.7 mg L-1 found in river water (WHO, 1996).

# 4. Estimation of Fe addition costs

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

# 5. Conclusions

In this study we demonstrated the potential of EC for the recovery of nutrients and other solids from food waste digestate. Fe was found to outperform Al in terms of COD removal from the soluble phase of food waste digestate. ~~Furthermore, Fe EC was successfully applied~~ ~~to enhance P in pressed cake solids from food waste digestate.~~ CFEC using 2.33 mM Fe and a 0.25 dilution of digestate was the optimal treatment tested. This treatment achieved the highest P recovery from the soluble phase of 22.4 mg-P per g-Fe added, the greatest reduction of solids in the filtrate (2.84 g per g-Fe added), substantial COD removal (of 254 mg L-1) and was estimated to be the least expensive ratio of Fe addition to digestate. ~~and was the this~~ ~~was achieved when CFEC used to apply 2.33 mM Fe to 0.25 dilution treatment was applied. It~~ ~~is worth noting that digestate is a complex matrix of components/contaminants and the~~ ~~amount of Fe dosed into the effluent is not exclusively removing P but rather a multitude of~~ ~~contaminants (e.g. metals, solids, COD).~~ Our results indicate that, in a scenario whereby recovered cake from EC-treated food waste digestate is applied to soil at a rate of 250 kg N per hectare per year, Cu, Ni, Zn, As, Cd, Cr, Mo and Pb would remain below their permitted application limits in the UK. Plant-growth trials are now required in further work to examine the quality of the nutrient-enhanced pressed cake solids from food waste digestion as a soil fertiliser for agricultural purposes.

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Conflict of Interest

## Declaration of interests

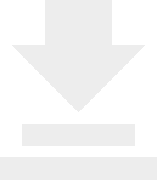
☐ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☒The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Dr Duarte Tito is the Technical Director of Elentec Limited. His company provided technical knowhow and bespoke electro-coagulation equipment to enable conduct of some research presented in the publication. Andrew P. Cooley is an employee at Elentec Limited.

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