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# 1 Valorisation of ferric sewage sludge ashes: potential as a 2 phosphorus source

3  
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11

## 12 Abstract

13 Sewage sludge ashes (SSA), although a waste, contain elements with socio-economic  
14 and environmental potential that can be recovered. This is the case of phosphorus (P).  
15 SSA from two Danish incinerators were collected during two years and characterized.  
16 The sampling was done immediately after incineration (fresh SSA) or from an outdoor  
17 deposit (deposited SSA). Although morphology and mineral composition were similar,  
18 physico-chemical and metal concentration differences were found between incinerator  
19 plants and sampling periods. No differences were observed between deposited and fresh  
20 SSA, except for the parameters directly influenced by disposal conditioning (e.g.  
21 moisture content). All the SSAs had high concentrations of P (up to 16 wt. %), but they  
22 all exceeded Danish EPA Cd and Ni thresholds for direct application at agricultural soil.  
23 Fresh and deposited SSA were acid washed aiming P extraction, achieving 50 g P/kg  
24 (approx. 37% of total P), but metals were also co-extracted to the liquid phase. To avoid  
25 and/or minimize the metals pollution of the extracted P, selective P recovery from the  
26 SSA was tested, using the electrodialytic (ED) process. ED laboratory cells, with 3  
27 compartments (3c) and 2 compartments (2c), and two acid concentrations (H<sub>2</sub>SO<sub>4</sub>, 0.08  
28 M and 0.19 M) were used for 7 days. The most concentrated acid solution increased P  
29 solubilization. The 2c-cell combined with the higher acid concentration resulted in  
30 higher P recoveries, 125 g of P/kg of SSA in the anolyte. The obtained results showed  
31 that the ED process is a valuable tool for the SSA valorisation as it promotes  
32 simultaneous P recovery and metals extraction from the SSA.

33

34 **Keywords:** *Electrodialytic process, acid extraction, characterization, phosphorus,*  
35 *metals, recovery*

## 36 1. Introduction

37 Incineration of sewage sludge ash (SSA) is a well-established technology (Werther and  
38 Ogada, 1999), being estimated that 300-400 kg of SSA are produced *per* ton of dried  
39 sewage sludge (Cyr et al., 2007). During the incineration process, organic contaminants  
40 and pathogens are destroyed, while achieving mass reduction, water evaporation and  
41 organic matter oxidation to NO<sub>x</sub>, SO<sub>x</sub> and CO<sub>2</sub>. After the incineration in a fluidized bed  
42 furnace, exhaust gas fines are collected by electrostatic precipitators or bag filters  
43 (Donatello et al., 2010b). Incineration cost is about 210-310 euros *per* ton of dry solids  
44 but some controversy arises as this technique promotes potentially hazardous SSA  
45 (Smith et al., 2009). In fact, incineration can concentrate up to 13 times the amount of  
46 metals when compared to the ones reported in the original matrix (Donatello et al.,  
47 2010b; Ottosen et al., 2013).

48 The SSA are generally considered a waste material to be disposed into landfill  
49 (Donatello et al., 2010b), but they can also be reused as adsorbents (Pan et al., 2003), in  
50 geotechnical applications (Ferreira et al., 2003) or in construction materials (Al Sayed et  
51 al., 1995; Anderson et al., 2002; Lin et al., 2005), since SSA is not considered  
52 hazardous waste (Donatello et al., 2010b). Due to the P concentration, typically between  
53 4 and 9% (Biswas et al., 2009; Franz, 2008), fly SSA can be applied at agricultural land,  
54 if it complies with the established metal thresholds (e.g. Danish EPA, Miljøstyrelsen).  
55 However, as the bioavailability of phosphorus in the SSA is low the untreated ash may  
56 not be a suitable fertilizer (Krüger and Adam, 2015; Herzel et al., 2016).

57 The SSA can be treated aiming P extraction (Donatello et al., 2010a; Franz, 2008;  
58 Weigand et al., 2012) using different technologies, e.g. thermal (Adam et al., 2009;  
59 Mattenberger et al., 2008; Mattenberger et al., 2010; Vogel et al., 2010), wet treatment  
60 process (Petzet et al., 2011; Petzet et al., 2012; Stark et al., 2006) or electrodialytic (ED)  
61 separation (Ebbbers et al. 2015; Guedes et al., 2014; Ottosen et al., 2014a). An  
62 experimental screening of ED P recovery and simultaneous metals removal has been  
63 conducted with a 3 compartment (3c) cell (Ebbbers et al., 2015; Guedes et al., 2014;  
64 Ottosen et al., 2014a), Figure 1a (Jensen and Villumsen, 1995), and 2 compartment (2c)  
65 cell (Ebbbers et al., 2015; Parés Viader et al., 2015), Figure 1b (Ottosen et al., 2014b). In  
66 the 3c cell, the possibility of P extraction from Al-rich and Fe-rich SSA was assessed  
67 using acid washing (Ottosen et al., 2013) and the ED process (Ottosen et al., 2014a).  
68 Phosphorus separation into the central compartment liquid and the anode compartment  
69 ranged between 15-85% for an Al-precipitated SSA and between 45-95% for a Fe-

70 precipitated SSA. The combination of ED process in the 2c-cell and initial acidification  
71 of the stirred suspension with mineral acids showed to be more effective in dissolving P,  
72 comparing to the 3c-cell. In the 2c-cell, removals up to 90% (Parés Viader et al. 2015)  
73 and 96% (Ebberts et al., 2015) were achieved in 7 days. A previous work reported  
74 phosphorus removals up to 70% have been achieved in the anode compartment of a 3c-  
75 cell for deposited and fresh Fe-rich SSA of the 3c-cell using 0.08M H<sub>2</sub>SO<sub>4</sub> (Guedes et  
76 al., 2014). However, neither the 2c-cell setup nor the influence of acid concentration  
77 were evaluated.

78 This work had two main objectives: (i) to evaluate the major differences between SSA  
79 collected in different sampling periods, storage conditions and incinerator plants  
80 (Lynettefællesskabet and Avedøre Spildevandscenter, Copenhagen, Denmark), and (ii)  
81 to assess P recovery from recently collected (fresh) and deposited SSA. This last  
82 objective was tested with SSA, collected in the incinerator at Lynettefællesskabet, using  
83 a combined acid washing and ED treatment with a 3c and 2c-cell setups.

84

85 **Please insert Figure 1**

86

## 87 **2. Experimental**

### 88 **2.1. Sewage sludge ashes**

89 The SSA samples were collected after incineration by fluidized bed combustion at  
90 Avedøre Spildevandscenter and Lynettefællesskabet wastewater treatment plants  
91 (WWTP), located in Copenhagen, Denmark. The plants treat waste water annually from  
92 345,000 and 500,000 population equivalent, respectively, with no major industrial  
93 discharges, prevailing non-industrial wastewater. At both facilities P in the wastewater  
94 is precipitated in a Bio-P tank followed by addition of iron salt. After incineration of the  
95 sewage sludge, the SSA is collected in electrofilters and the ferric SSA is disposed of in  
96 separate landfills for SSA only. In total, 8 samples of SSA were collected. From  
97 Avedøre, two samplings (2012 and 2014) were made directly after the electrofilters  
98 (fresh). From Lynettefællesskabet, six samples (twice in 2012 and once in 2013) were  
99 collected from the electrofilters (fresh) and at the disposal site (deposited). The  
100 deposited SSA were collected from the top part of the SSA pile in an open air deposit,  
101 and the storage time is unknown. After sampling, the SSA was stored in closed plastic  
102 containers at room temperature until the experimental work was carried out.

103

## 104 **2.2. SSA characterization**

105 Characterization of the SSA and metals extraction were carried out with oven dried SSA  
106 (105 °C). For all characterization procedures five replicates of each type of SSA were  
107 used. Water content of the SSA was calculated as weight loss at 105 °C for 24 h. The  
108 pH was measured in a 1:2.5 (mass:volume) suspension in distilled water using a  
109 Radiometer pH electrode. Loss on ignition (LOI) was determined in a muffle at 550 °C  
110 for 30 min. The concentrations of different elements were measured after pre-treatment  
111 of the SSA in accordance to DS259 (2003), as 1.0 g of SSA and 20.0 mL (1:1) HNO<sub>3</sub>  
112 extracted at 200 kPa, 120 °C for 30 min and then vacuum filtered through a 0.45 µm  
113 filter. Phosphorus, Al, Fe, Zn, Cu, Pb, Cr, Cd and Ni were measured by inductively  
114 coupled plasma optical emission spectrometry (ICP-OES).

115 Morphology analysis was performed by scanning electron microscopy supported by  
116 energy dispersive spectrometry (SEM-EDX), without pre-treatment of the SSA samples  
117 (3 replicates were analysed for each SSA). The accelerating voltage of the SEM was 20-  
118 25 kV with large field detector (and X-ray cone). Different areas of the samples were  
119 analysed by SEM and the element distribution was examined using EDX. SSA  
120 mineralogy was studied by X-ray diffraction (XRD) for the identification of the major  
121 crystalline phases. The instrument was a PANalytical X'Pert Pro operating at 45 kV and  
122 40 mA applying Cu K $\alpha$  radiation with a 2 $\Theta$  X'Celerator detector. The samples were  
123 scanned in the range of 4-70 2 $\Theta$ . The diffractograms were interpreted by using the  
124 ICDD PDF-4 database for minerals.

## 125 **2.3. Relation between pH release of metals and P**

127 To determine the pH dependent extraction of P and metals, 10 g dried SSA were  
128 suspended in 50 ml HNO<sub>3</sub> of different concentrations (0.00; 0.01; 0.05; 0.08; 0.10; 0.30;  
129 0.50; 0.70 and 1.0 M). The suspensions were placed at an agitating table for 7 days at  
130 room temperature. At the end of the experiment, pH was measured and suspensions  
131 were filtered by vacuum using 0.45 µm filters and the concentrations determined by  
132 ICP-OES. Each extraction was made in duplicate and reference samples were made  
133 using distilled water with the same procedure. pH dependent extractions was carried out  
134 for the Lynettefællesskabet SSA (2012, 1<sup>st</sup> sampling).

## 135 **2.4. ED treatment of SSA**

137 Two ED laboratory cells were used (Figure 1). The cells were made of plastic with an  
138 internal diameter of 8 cm, with a suspension compartment length of 10 cm and the  
139 electrode compartment of 5 cm. Electrode compartments were separated from the  
140 suspension compartment by an anion-exchange membrane and/or a cation-exchange  
141 membrane from Ionics (anion exchange membrane, AEM - AR204 SZRA B02249C  
142 and cation exchange membrane, CEM - CR67 HUY N12116B). The electrodes were  
143 platinum coated titanium from Permascand. A power supply (Hewlett Packard E3612A)  
144 was used, maintaining a constant current of 50 mA, corresponding to a current density  
145 of 0.08 mA/cm<sup>2</sup>, whereas in the 2c-cell was of 0.10 mA/cm<sup>2</sup>. The SSA was stirred in the  
146 suspension compartment by a flexible plastic flab, coupled to a HETO motor, with a  
147 rotation velocity rate of up to 1300 rpm. The SSA was suspended in H<sub>2</sub>SO<sub>4</sub> at 0.19 M in  
148 the 3c-cell and 0.08 and 0.19 M for the 2c-cell, in the ratio of 1:10 (mass:volume), and  
149 500 mL of 0.01 M NaNO<sub>3</sub> was used as electrolyte, with pH adjusted to 2 with 1:1  
150 HNO<sub>3</sub>. A total of six ED experiments with SSA from Lynettefællesskabet (2012, 1<sup>st</sup>  
151 sampling), two experiments for each SSA sample (fresh and deposited) were carried out  
152 for 7 days with different experimental conditions (Table 1).

153 During the experiments voltage, current, pH in the electrolytes and pH and conductivity  
154 in the SSA suspension were measured. The pH of the electrolytes was manually  
155 adjusted to pH between 1 and 2 with 1:1 HNO<sub>3</sub> when necessary.

156

157 **Please insert Table 1**

158

159 At the beginning and at the end of each experiment, samples were taken from  
160 electrolytes and from the suspension, for P and metal analyses. At the end of the  
161 experiments the SSA suspensions were drained through filter paper (grade 113, pore  
162 size 30 µm) before drying, to separate the solids from the liquid phase, and water  
163 content was measured. Membranes and cathode were soaked in HNO<sub>3</sub> (1 and 5 M,  
164 respectively) for 24 hours to release metals for further analysis. The SSA (three  
165 replicates), aqueous phases, electrolytes, membranes and cathode were analysed for P  
166 and metal contents, using the same ICP-OES.

167 The removal percentages were calculated by dividing the mass of element removed to  
168 different parts of the cell (electrodes, electrolyte solutions and membranes) by the sum  
169 of mass found in the different parts of the cell at the end of the experiment.

170

## 171 *2.5 Minteq calculations*

172 Visual MINTEQ 3.1 was used to gain further insight into the formation of charged  
173 phosphorus complexes which could be transported through the AEM and CEM. For the  
174 calculations the chemical composition of the ash liquid solution in experiments after the  
175 7 days with elements present in SSA with Fe as  $\text{Fe}^{3+}$  or as  $\text{Fe}^{2+}$  was used. The pH was  
176 fixed to the final experimental pH and temperature to 25 °C. The Ionic strength was  
177 calculated in the program.

## 178 *2.6. Statistical analysis*

179 Physico-chemical characteristics, element concentrations in all fresh and deposited SSA  
180 were subjected to analysis of variance (ANOVA) and then compared by Tuckey  
181 multiple range test at  $p < 0.05$ . A Student t-test was used for comparison of SSA  
182 collected in the same incinerator plant.  
183

# 184 **3. Results and Discussion**

## 185 *3.1. Chemical characterization of different SSA*

186 The characterization of fresh Avedøre SSA (2012 and 2014) and fresh/deposited  
187 Lynettefællesskabet SSA (2012 and 2013) is presented in Table 2.

188 Regarding physico-chemical characteristics, the changes in pH and conductivity did not  
189 follow a specific tendency, differences being attributed to SSA heterogeneity  
190 (wastewaters from different urban areas in Denmark but with no major industrial  
191 discharges). The high difference in water content between deposited (up to 16%) and  
192 fresh ( $\leq 0.1\%$ ) SSA was related to storage (e.g. air humidity, rain). The LOI was  
193 generally low ( $< 0.9\%$ ), only increasing for one Lynettefællesskabet sample (2.7%).  
194 Both plant facilities have thus shown sufficient incineration conditions.  
195

## 196 **Please insert Table 2**

197  
198  
199 The concentration of the different elements was in the same range in all studied SSA.  
200 The P content was generally high in all SSA (between 7 and 16 wt. %). Differences in P  
201 are majorly attributed to population diet (Mihelcic et al., 2011), type of wastewater  
202 treatment plant system, and type of detergents used. In general, the Zn concentration  
203 was higher in Lynettefællesskabet 2012 SSA than in the other samples. In 2014,  
204 Avedøre Spildevandscenter had higher concentrations of Cu and Pb than in 2012 or in

205 Lynettefællesskabet. Again, these variations can be related to the characteristics of the  
206 area covered by the WWTP, which directly influence the heterogenous characteristics of  
207 the daily SS flows. Most of the studied Danish SSA were in accordance with reported  
208 literature ranges (Donatello et al., 2010a; Ebbers et al. 2015; Franz, 2008; Levlin et al.,  
209 2003; Ottosen et al., 2013) (also in Table 2, for comparison purposes), except for some  
210 samples with higher Zn, Cu and Pb values or lower Fe and Ni concentrations. In this  
211 work, Cr levels were below all the concentrations reported in literature.

212 The Danish EPA has two set of limiting values for metals when spreading waste at  
213 agricultural land. The dry matter related concentrations (see Table 1) met the limiting  
214 values except for Cd (2.2-3.5 mg/kg) and Ni (35-62 mg/kg) for all SSA, and Pb for one  
215 sampling period in Lynettefællesskabet and Avedøre (Table 2). As previously  
216 discussed, the pre-treatment of the SSA would significantly improve the amount of P  
217 bioavailable for agricultural application.

218

### 219 **3.2. Study of SSA from Lynettefællesskabet (2012)**

220 Deposited and fresh Lynettefællesskabet SSA from the first 2012 sampling (Table 2)  
221 were further analysed, including morphology, mineral composition and potential of acid  
222 washing to remove P. These SSA were chosen to deepen the comparison between fresh  
223 and deposited SSA.

224

#### 225 **3.2.1. Morphology and mineral composition**

226 SEM analysis show that the SSA particles generally had irregular shaped (Figure 2) and  
227 no major differences were found between fresh and deposited samples. Additionally,  
228 SEM/EDX analysis revealed that O, P, Fe, Al, K, Na, Mg, Si, S, and Ca were  
229 distributed all over the particle surfaces in both SSA.

230

#### 231 **Please insert Figure 2**

232

233 The mineralogy of fly SSA generally represents an assemblage of crystalline and  
234 amorphous phases (Bayuseno and Schmahl, 2011). The major mineral phases identified  
235 by XRD are presented in Figure 3 and show differences between deposited and fresh  
236 SSA. Silicon oxide in the form of quartz ( $\text{SiO}_2$ ) was identified in both SSA, agreeing  
237 with published data for other SSA (Cyr et al., 2007) where Si is likely to be present in  
238 its crystalline or amorphous phase. Phosphates on both SSA were indicated as calcium



239 phosphates, but the only chemical formula clearly identified was as whitlockite  
240  $[\text{Ca}_9(\text{MgFe})(\text{PO}_4)_6\text{PO}_3\text{OH}]$ . Hematite ( $\text{Fe}_2\text{O}_3$ ) was also identified, being the iron oxide a  
241 consequence of SS incineration (800-900 °C) in excess air which promotes the existence  
242 of metals as oxides (Donatello et al., 2010b).

243

244 **Please insert Figure 3**

245

246 There was  $\text{CaSO}_4$  in both samples, however difficult to completely identify. In the fresh  
247 sample, it is likely present as anhydrite ( $\text{CaSO}_4$ ) and in the deposited, it was not clear,  
248 although not anhydrite nor gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ). However, anhydrite can be present  
249 in SSA when they are fresh but as a result of “hydration” of the SSA, gypsum may start  
250 to form (Bayuseno and Schmahl, 2011), though this was not confirmed by XRD in the  
251 present work. In SEM/EDX a S rich region was observed and can belong to i) an  
252 amorphous  $\text{CaSO}_4$  or ii)  $\text{Na}_2\text{SO}_4/\text{K}_2\text{SO}_4/\text{NaKSO}_4$  condensed on this Ca-rich particle.

253 The amorphous phase in other SSAs is between 40-74% (Cyr et al., 2007). In this case,  
254 from XRD patterns of fresh and deposited SSA samples it can be inferred that some  
255 amorphous phases are present due to the slight hump between  $2\theta$  of 10 and  $30^\circ$  but the  
256 material is not prevalently amorphous.

257 A semi-quantitative method, Reference Intensity Ratio (RIR), based on the diffraction  
258 intensity of the strongest peak of a given phase in comparison with corundum, was used  
259 to indicate the relative mineral mass fractions in the two SSA. The RIR method  
260 indicated that the mass distribution of the major crystalline phases in both SSA were as  
261 following: calcium phosphate and quartz > iron oxide and calcium sulphate.

262

### 263 ***3.2.2. Acid washing of SSA***

264 The percentage of P extracted from the SSA as a function of pH is shown in Figure 4.  
265 Storage did not affect the extent of P extraction even at the strongest concentration, 1.0  
266 M, where more than 50 g/kg were extracted. This 40% of total P mobilization is in  
267 accordance with the average percentage reported in other works, for the same pH  
268 (Ottosen et al., 2013). A pH decrease, other L/S ratios or extracting agents could have  
269 increase the total amount of mobilized P (Ottosen et al., 2013).

270 Regarding storage, the release of a specific element as a function of pH is very similar  
271 for fresh and deposited SSA, except for a slight variation of Cd at pH between 2 and 3.

272 The major mineralogy of the material and the phases controlling the solubility of major

273 trace elements (as suggested by the results of mineralogy test) were also not affected,  
274 neither the amount of elements extracted. Instead, extraction is mainly correlated with  
275 pH changes, particularly at low pH values. At low pH values the solubilized percentages  
276 were: Zn 65%, Cu approx. 55%, and less than 33% Ni and Al (Figure 4). Iron was  
277 bound strongly in both SSA, and even at the lowest pH, less than 2% was solubilized.  
278 This is in accordance to what was expected as  $\text{Fe}_2\text{O}_3$ , which is insoluble in acid  
279 (Donatello et al., 2010b), was identified in larger quantities in both SSA. The XRD  
280 investigation also showed the presence of  $\text{Ca}_9(\text{MgFe})(\text{PO}_4)_6\text{PO}_3\text{OH}$  in the SSA. But,  
281 since at low pH, only a small amount of Fe was extracted in contrast to more than 35%  
282 of P in both samples (Figure 4) these two elements may have not been associated in the  
283 SSA, even though Fe was originally used for precipitation of P during the wastewater  
284 treatment. As metals mobilization started approximately at the same pH than P  
285 mobilization, the obtained acidic liquid solution was a mixture between P and metals,  
286 and a further separation step is needed.

287

288 **Please insert Figure 4**

289

290 **3.2.3. Electrodialytic treatment**

291 **3.2.3.1. Mass balance**

292 Mass balance of an element was defined as the relation between the sum of mass found  
293 in the different parts of the cell at the end of the experiment and the initial mass  
294 calculated on basis of the mean initial concentration. The mass balances for the  
295 elements presented high variability for all the experiments. An inhomogeneous  
296 distribution of metals in the ash and, consequently, imperfect mass balances are  
297 expected when working with these type of samples. Finally, even though careful  
298 handling of all samples the precipitation of insoluble compounds may occur in e.g. set-  
299 up tubing. Mass balance in the different experiments varied between 80–120% with no  
300 linear tendency.

301

302 **3.2.3.2. Electrodialytic P-separation**

303 Table 3 shows the initial conditions and the changes in pH, conductivity and mass loss  
304 after the ED experiments. Before the experiment the SSA was suspended in sulphuric  
305 acid (0.08 and 0.19M) aiming to convert the precipitated and adsorbed P into mobile

306 ionic forms able to electromigrate during the ED treatments. The initial pH in the  
307 central cell compartment varied between 1.0 and 2.6 for the SSA (Table 2). During the  
308 ED experiments, pH in the SSA compartment slightly decreased in the 3c-cell and  
309 continued to decrease in the 2c-cell due to the generation of  $H^+$  in the anode  
310 compartment. Water splitting at the AEM may have also contributed for the  
311 acidification. The dissolution of ash particles during the ED treatment results in a mass  
312 loss that can be quite significant (Guedes et al., 2014; Ottosen et al., 2006) as it can be  
313 seen in Table 3. In the 2c-cell experiments there was a conductivity increase at the end  
314 of the experiments. This is due to the electrolysis at the anode and solubilisation of SSA  
315 ions that are not transported out of the suspension as fast as produced. In the 3c-cell the  
316 solubilized ions from the SSA suspension are migrating either to the anode or cathode  
317 compartments. Consequently, the central compartment is being depleted from ions,  
318 which decreases the conductivity of the suspension.

319

320 **Please insert Table 3**

321

322 In the ED cells, the released metals are expected to be found as positively charged  
323 species whereas the solubilised P is more likely to be found in negatively charged or  
324 neutral species at acidic pH ( $H_3PO_4$ ,  $H_2PO_4^-$ ), except for Al-P, Ca-P and Fe-P  
325 complexes formation. In the 3c-cell the main objective is to dissolve and mobilize the P  
326 from the SSA suspension (central compartment) to the anode compartment and the  
327 metals to the cathode compartment. In the 2c-cell the aim is to dissolve P and metals  
328 from the SSA keeping P in the suspension (anolyte) whereas the metals are removed  
329 into the cathode compartment. Afterwards the ED treatment using the 2c-cell, the  
330 suspension (anolyte) is filtered, being the P rich filtrate cleaned from ash particles.

331 In this study, a more concentrated  $H_2SO_4$  solution was used in the 3c-cell compared to  
332 the previous work where 0.08 M solution was used (Guedes et al., 2014). The increase  
333 in the acid concentration improved P solubilization by 17% (experiments F-3-0.19 and  
334 D-3-0.19, Figure 5). In total between 24.9 and 29.4% of P were removed to the anolyte  
335 of the 3c-cell, mainly as  $H_2PO_4^-$  (Table 4), whereas in the previous work 17.9 and  
336 18.8% were removed to the anolyte after 7 days (Guedes et al., 2014).

337

338 **Please insert Figure 5**

339

340 When the 2c-cell with 0.08 M of H<sub>2</sub>SO<sub>4</sub> was used 91% of P for F-2-0.08 and 67% for  
 341 D-2-0.08 were solubilized. From these, 14 and 11% of P (F-2-0.08 and D-2-0.08,  
 342 respectively) electromigrated to the cathode compartment, probably due to the  
 343 complexation with Al, Fe and Ca forming positively charged species like AlHPO<sub>4</sub><sup>+</sup>,  
 344 CaH<sub>2</sub>PO<sub>4</sub><sup>+</sup> and FeH<sub>2</sub>PO<sub>4</sub><sup>2+</sup> (Table 4). At the end, the anolyte solution still contained  
 345 77% of P in F-2-0.08 and 56% in D-2-0.08 (Figure 5) being mainly present as H<sub>3</sub>PO<sub>4</sub>  
 346 and H<sub>2</sub>PO<sub>4</sub><sup>-</sup> (Table 4). In the acid washing test, this difference in P solubilization was  
 347 not observed. One hypothesis is that deposited SSA sample used in the experiment D-  
 348 2-0.08 may have contained more acid-insoluble P-complexes (samples heterogeneity)  
 349 influencing the results. Still, in these two experiments, the type of SSA influenced the  
 350 total P solubilization by the different setups and, consequently, the amount collected in  
 351 the anolyte solution. In this case, the best option may be to use ash collected  
 352 immediately after incineration. The increase in the acid concentration in the 2c-cell set-  
 353 up resulted in higher P solubilization from the SSA, approx. 99% in both cases. From  
 354 these, 93 and 97% in the F-2-0.19 and D-2-0.19, respectively, remained in the anolyte  
 355 as H<sub>3</sub>PO<sub>4</sub> and H<sub>2</sub>PO<sub>4</sub><sup>-</sup> (Table 4). Only 7% in F-2-0.19 and 2% in D-2-0.19  
 356 electromigrated toward the cathode compartment mainly as Al, Ca, and Fe complexes  
 357 (AlHPO<sub>4</sub><sup>+</sup>, Al<sub>2</sub>PO<sub>4</sub><sup>+3</sup>, CaH<sub>2</sub>PO<sub>4</sub><sup>+</sup>, FeH<sub>2</sub>PO<sub>4</sub><sup>2+</sup> and FeHPO<sub>4</sub><sup>+</sup>, Table 4).

358 In terms of metals, generally, higher removals were achieved when 0.19M of H<sub>2</sub>SO<sub>4</sub>  
 359 was used compared to the use of 0.08M, independently of the cell design (3c-cell or 2c-  
 360 compartment). The metals solubilization after the 7 days of experiment were:

- 361 • Fe (9%) < Pb (14%) < Cr (16%) = Ni < Zn (56%) < Al (57%) < Cu (69%) < Cd  
 362 (82%) for F-3-0.19
- 363 • Pb (10%) < Fe (12%) < Cr (20%) < Ni (36%) < Al (44%) < Zn (73%) < Cu  
 364 (76%) < Cd (82%) for D-3-0.19
- 365 • Pb (7%) < Fe (10%) < Cr (11%) < Ni (16%) < Cd (18%) < Al (53%) < Cu  
 366 (55%) < Zn (64%) for F-2-0.08
- 367 • Pb (9%) < Fe (12%) = Cr < Ni (17%) < Cd (23%) < Al (42%) < Cu (44%) < Zn  
 368 (51%) for D-2-0.08
- 369 • Fe (6%) < Cr (10%) < Pb (12%) < Ni (13%) < Cd (23%) < Al (53%) = Zn < Cu  
 370 (69%) for F-2-0.19
- 371 • Fe (8%) < Pb (14%) < Cr (16%) < Ni (18%) < Cd (27%) < Zn (51%) < Al  
 372 (61%) < Cu (64%) for D-2-0.19

373 The exception was Cd which solubilization was higher in the 3c-cell, 82% in both  
374 experiments (F-3-0.19 and D-3-0.19) contrasting to the 23 and 27% in the F-2-0.19 and  
375 D-2-0.19, respectively. In the 2c-cell experiments, between 73-82% of Cd remained in  
376 the ash whereas in the 3c-cell the value was 18%. Although unclear, a possible  
377 explanation is that the reactions that occurred in the anode end, where the SSA is  
378 placed, might have affected the Cd solubilization (e.g. precipitation) and this should be  
379 further investigated. Still, higher solubilization values for Cd (89%) were obtained by  
380 Ebbers et al. (2015) using a 2c-cell with 0.19 M of H<sub>2</sub>SO<sub>4</sub>.

381 As Fe and Pb are tightly bounded to more insoluble ashes particles (Ottosen et al.,  
382 2014a) neither the increase in the acid concentration nor the change in the cell design  
383 improved their removals.

384 Comparing to the previous work (Guedes et al., 2014) generally, higher percentages  
385 were solubilized from the SSA in these experiments (F-3-0.19 and D-3-0.19) due to the  
386 increased acid concentration, 0.19 M. Between more 3% (Fe in the fresh SSA) and 36%  
387 (Cd in the deposited SSA) were solubilized from the SSA comparing to the results  
388 obtained with 0.08 M (Guedes et al., 2014). No differences were found for Al in the  
389 deposited SSA (D-3-0.19) and Cr and Cu in the fresh SSA (F-3-0.19).

390

391 **Please insert Table 4**

392

### 393 ***3.2.5. Techniques comparison***

394 At the end of all ED experiments pH was between 1.9 and 1.2. Through comparison to  
395 acid washing, it was expected that at least 35% of P had been solubilized from the SSA  
396 in the ED cells (Figure 4) staying in the anode compartment in the 2c-cell or  
397 electromigrating to anode compartment in the case of the 3c-cell. In all cases, the  
398 amount of P solubilized was higher in the ED cell (generally higher than 80%, except in  
399 D-2-0.08 that was 67%) than in the acid washing. This may be attributed to four factors:  
400 (i) the use of sulphuric acid in the ED treatment, instead of the nitric acid used in the  
401 acid washing; (ii) the combination of an increased solubilisation of P compounds, due to  
402 equilibrium displacement by Al, Ca and Fe cations migration into the catholyte (Parés  
403 Viader et al., 2015); (iii) a higher liquid-to-solid ratio (L/S) in the ED remediation  
404 (L/S=10) than in the acid extraction experiments (L/S=5); and (iv) the continuous  
405 generation of H<sup>+</sup> ions (either by water electrolysis or water splitting). The main

406 advantage of the ED process comparing to acid washing is the simultaneous extraction  
407 and separation of P from the other element in the ash, namely the metals.  
408 Between cell designs, the use of a 2c-cell setup increased overall efficiency of P  
409 recovery after 7 days of experiment, under the conditions here tested. Although the  
410 applied current was the same (50 mA) the corresponding current density was different  
411 ( $0.08 \text{ mA/cm}^2$  in the 3c-cell vs  $0.10 \text{ mA/cm}^2$  in the 2c-cell) which may have influenced  
412 the results. Also, P dissolution seems to be faster and more complete in the 2c-cell  
413 experiments than in the 3c-cell, possibly due to the faster formation of  $\text{H}^+$  ions due to  
414 water electrolysis. Another important factor is that, at the end of experiments, the P that  
415 stays in the suspension of the anode compartment (anolyte) in the 2c-cell (either as  
416 uncharged species or anions) is accounted as recovered whereas in the 3c-cell the  
417 amount of P that stays in the central compartment liquid (as neutral species) is not  
418 considered recovered. At the end, and though the amount of P solubilized in the 3c-cell  
419 and 2c-cell using 0.19 M of  $\text{H}_2\text{SO}_4$  was very similar (approx. 92% and 99%,  
420 respectively), the P recovered was only 25-29% in the 3c-cell contrasting to the 93-97%  
421 in the 2c-cell. Still, in the 2c-cell set-up, the P was not completely separated from the  
422 metals as it can be seen in Table 5 (for example, 32% of Al for F-2-0.19 and 45% for D-  
423 2-0.19 are still in the anolyte). From the total amount of elements analysed, the anolyte  
424 solution of D-2-0.19 was constituted by 90% of P and 10% of metals, whereas F-2-0.19  
425 anolyte contained 93% of P and the remaining 8% were metals.

426

427 **Please insert Table 5**

428

#### 429 **4. Conclusions**

430 The SSA from Avedøre and Lynettefællesskabet incinerator plants showed no large  
431 differences between their chemical characteristics, except for those related with SSA  
432 heterogeneity. Additionally, samples collected at different stages (fresh and deposited)  
433 at the same incineration plant showed no trending differences in metal content and  
434 physico-chemical parameters. The exception was the moisture content, which was  
435 directly related with the disposal conditions.

436 All the SSA exceeded Danish EPA Cd and Ni thresholds regarding agriculture  
437 application. Their high P concentration (up to 16 wt. %) justifies the need of P recovery.  
438 More than 50 g of P/kg was extracted from fresh and deposited Lynettefællesskabet  
439 SSA (2012) by acid washing, together with a high amount of metals.

440 The ED process proved to be a viable option for simultaneous extraction and separation  
441 of metals and P thus promoting the valorization of the SSA. The use of a 2c-cell to  
442 solubilize and separate P from the metals seems to be the best option with the best  
443 results being achieved when 0.19 M of H<sub>2</sub>SO<sub>4</sub> were used (up to 99% of P  
444 solubilization). In total, approx. 125 g of P/kg of SSA were recovered in the analytes  
445 showing that the SSA are valuable P sources. However, the optimization of the ED  
446 conditions, current and duration, are still needed aiming at decreasing energy costs.  
447 Although more tests are needed, the type of ash used in the process may influence P  
448 recovery, and it needs to be taken into account. In this study, the SSA collected after the  
449 incineration allowed to recover more 24% of P than the use of the SSA collected from  
450 the deposit, when the 2c-cell with 0.08 M of sulfuric acid was used.

451

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462

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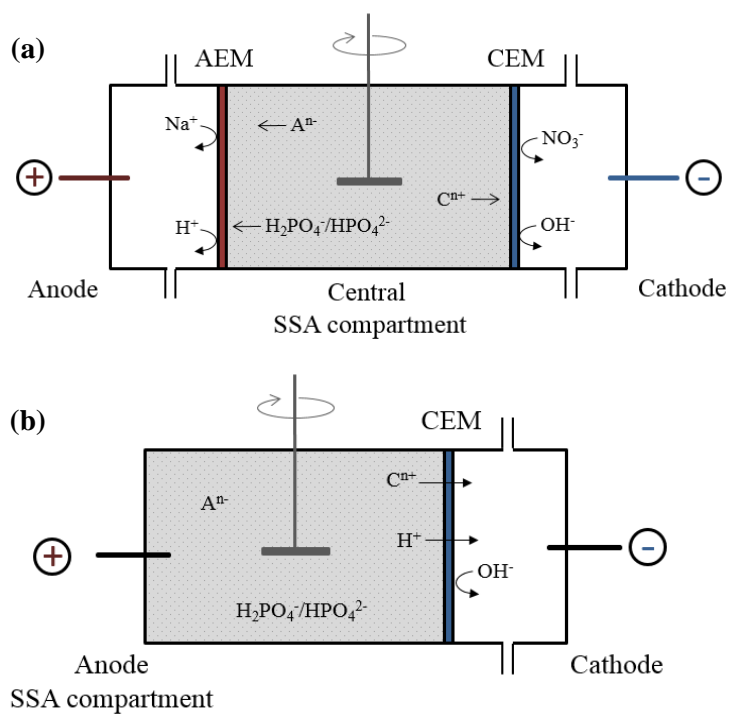
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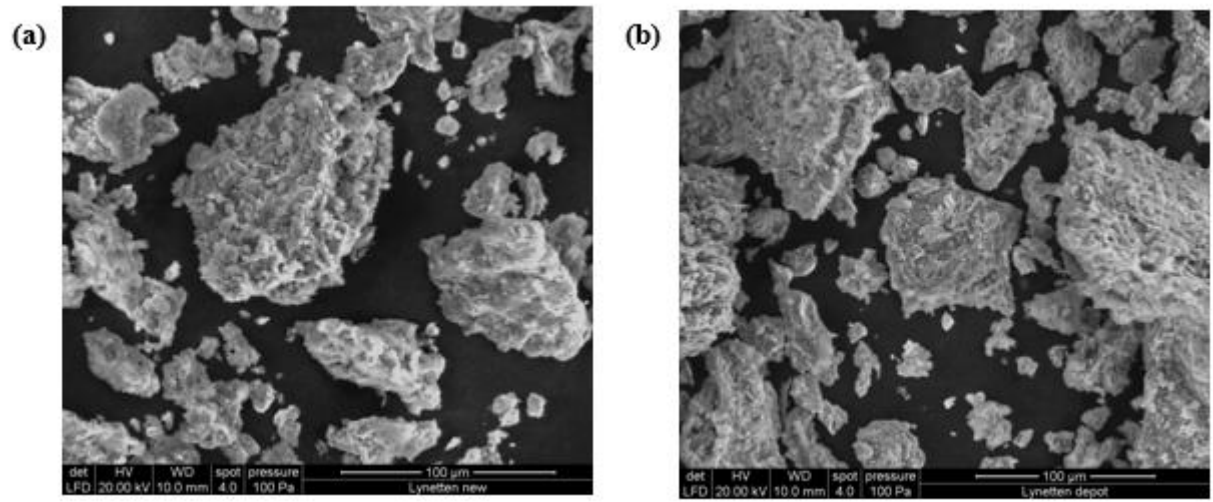
**Highlights**

- Sewage sludge ashes fresh and deposited were studied aiming phosphorus recovery
- Ashes physico-chemical differences depended on incinerator plant and sampling date
- Phosphorus solution after acid wash contained high amount of heavy metals
- The 2 compartment ED-cell improved phosphorus extraction and recovery
- A cleaner phosphorus solution was obtained after ED

Figure 1

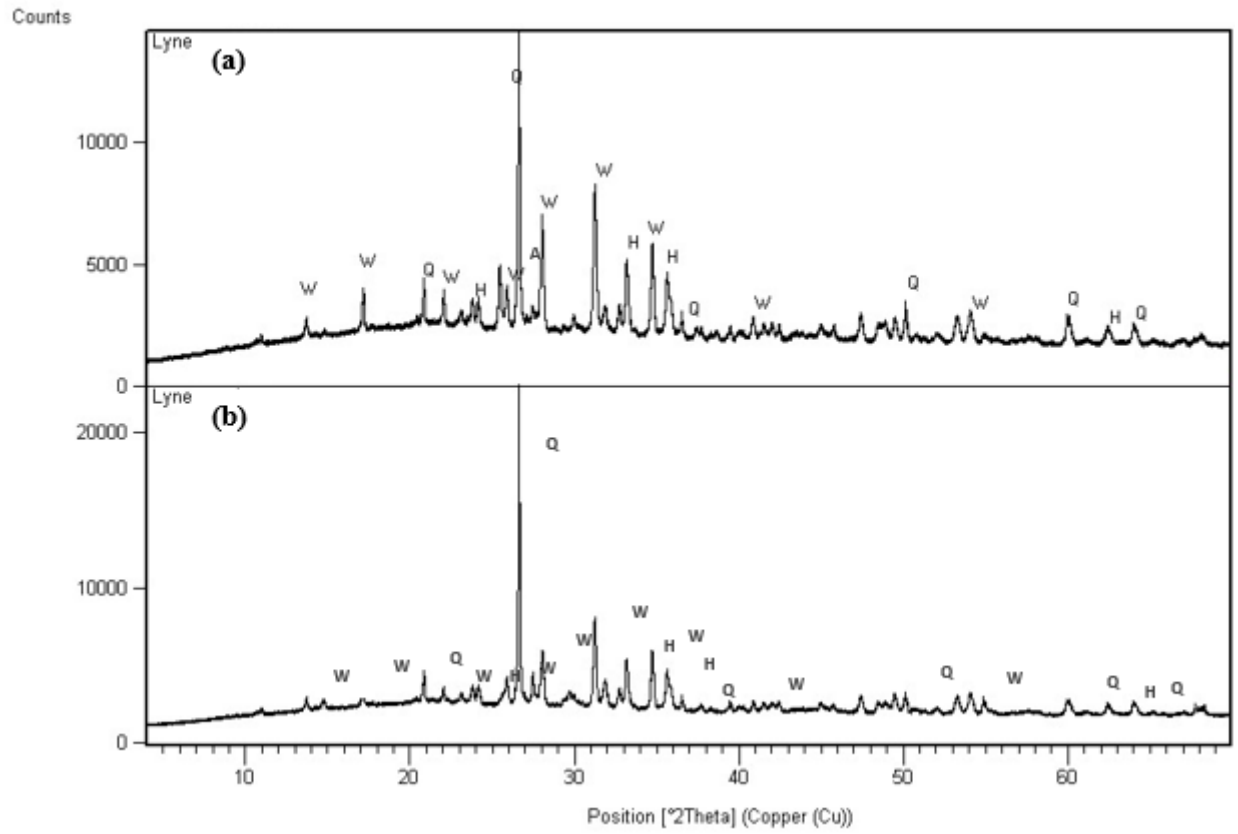


**Figure 1.** Schematic view of (a) 3-compartment and (b) 2-compartment electrodialytic laboratory cell. (CEM: cation exchange membrane; AEM: anion exchange membrane)



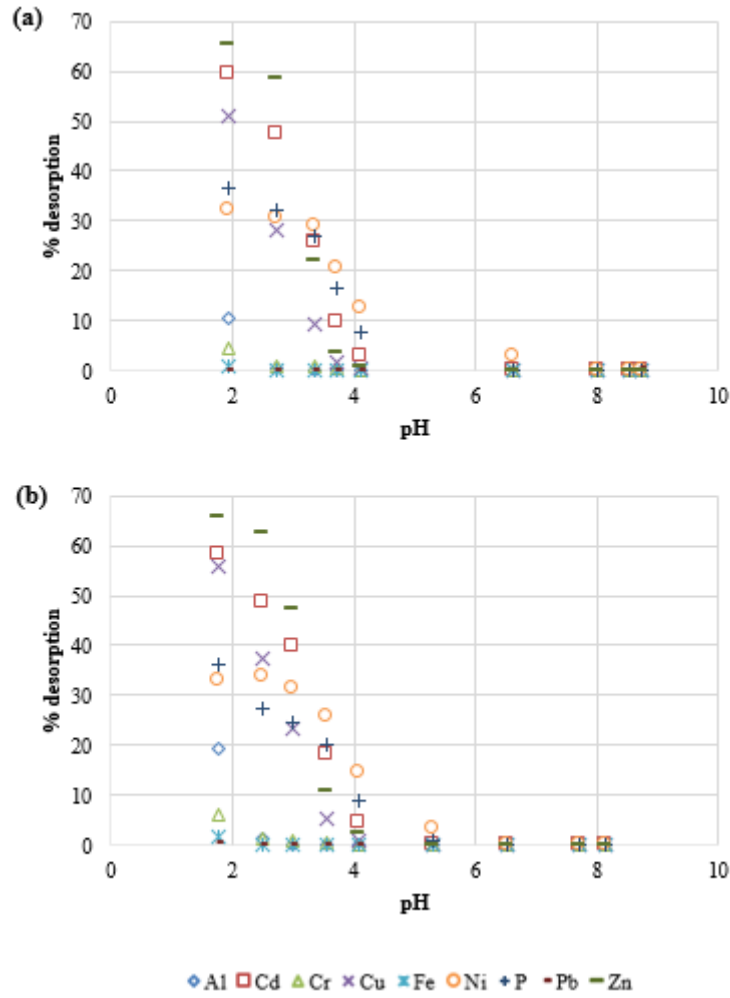
**Figure 2.** SEM micrographs of (a) fresh and (b) deposited Lynettefællesskabet SSA 2012 1<sup>st</sup> sampling.

Figure 3



**Figure 3.** XRD diffractograms of (a) fresh and (b) deposited Lynettefællesskabet SSA 2012 1<sup>st</sup> sampling (Q - Quartz, W- Whitlockite, H – Hematite, A – Anhydrite).

Figure 4



**Figure 4.** Phosphorus and metal extraction as a function of pH in batch extraction experiments on (a) fresh and (b) deposited SSA Lynettefællesskabet 2012 (1<sup>st</sup> sampling) using HNO<sub>3</sub>.

Figure 5



**Figure 5.** Percentage of heavy metals and phosphorus in the different matrices after the ED treatment in the 3C and 2c-cell design.

**Table 1.** Experimental design for the Lynettefællesskabet SSA (2012, 1<sup>st</sup> sampling).

<b>Experiment</b>	<b>Lynetten SSA sample</b>	<b>Cell design</b>	<b>SSA compartment</b>	<b>H<sub>2</sub>SO<sub>4</sub> (M)</b>
<b>F-3-0.19</b>	Fresh (2012)	3 compartment	central	0.19
<b>D-3-0.19</b>	Deposited (2012)			
<b>F-2-0.08</b>	Fresh (2012)	2 compartment	anode	0.08
<b>D-2-0.08</b>	Deposited (2012)			
<b>F-2-0.19</b>	Fresh (2012)			0.19
<b>D-2-0.19</b>	Deposited (2012)			



Table 2

**Table 2- Characteristics of the studied Danish SSA including heavy metal concentrations in relation to the total dry mass (mean ± STD) and limiting values for spreading at agricultural land. Values reported in literature were also used for comparison.**

Parameter	Lynettefællesskabet						Avedøre		Limiting values (mg/kg) <sup>a</sup>	Literature <sup>b</sup>
	2012 (1 <sup>st</sup> sampling)		2012 (2 <sup>nd</sup> sampling)		2013		2012	2014		
	Fresh <sup>(1)</sup>	Deposited <sup>(1)</sup>	Fresh	Deposited	Fresh	Deposited	Fresh	Fresh		
Physical and chemical characteristics	pH (H <sub>2</sub> O)	12.44 ± 0.01 *#	8.85 ± 0.03 *	12.4 ± 0.05 *#	8.3 ± 0.03 *	12.6 ± 0.02 #	12.6 ± 0.01 #	10.4 ± 0.0 *	9.6 ± 0.10 *	
	Water content (%)	0.10 ± 0.18 *	16 ± 0.38 *	0.09 ± 0.03	14.4 ± 0.09	3.70 ± 0.04 #	3.93 ± 0.07 #	-	0.16 ± 0.01	
	Conductivity (mS/cm)	3.23 ± 0.51 *	4.81 ± 0.13 *#	7.81 ± 0.02 *	5.59 ± 0.11 *#	10.54 ± 0.21 #	9.97 ± 0.82 #	2.12 ± 0.02 *#	2.52 ± 0.14 *#	
	Loss on ignition (550 °C; %)	0.15 ± 0.05 *#	0.92 ± 0.08 *	0.25 ± 0.05 *#	0.16 ± 0.00 *	2.58 ± 0.01 *	0.47 ± 0.04 *	0.3 ± 0.1 *#	0.57 ± 0.07 *	
	Solubility in water (%)	1.8 ± 0.1 *	3.1 ± 0.0 *	5.12	0.84	5.3 ± 0.0	5.6 ± 0.79	-	29.5	
	Gas production (mL gas/g)	1.6 ± 0.1	1.9 ± 0.2	-	-	-	-	-	-	
Elements	P (g/kg)	134 ± 1	130 ± 5	161 ± 12	-	72.5 ± 0.01 *	68.3 ± 1.5 *	112 ± 2 #	105.4 ± 3.8 #	39-123
	Al (g/kg)	22.6 ± 0.5 *	21.5 ± 0.7 *	21.2	19.5	14.2 ± 0.0	-	20.3 ± 0.5 *	22.1 ± 0.6 *	19.7-218
	Fe (g/kg)	60.0 ± 1.4	62.0 ± 2.3	44.0	-	36.3 ± 0.1	-	78.2 ± 2.9 *	53.2 ± 1.5 *	47-200
	Zn (mg/kg)	3335 ± 77 *#	3157 ± 128 *	3060 ± 222 #	2810 ± 117	2414 ± 10 *#	2270 ± 66.7 *#	2160 ± 60 *#	2410 ± 72 *#	<b>4000</b> 448-2737
	Cu (mg/kg)	758 ± 4.9 *	733 ± 9 *	711 ± 5.65 *	694 ± 6.49 *	512 ± 4 #	507 ± 7.58 #	550 ± 10 *	815 ± 18 *	<b>1000</b> 417-690
	Pb (mg/kg)	293 ± 44 #	297 ± 9	102 ± 2.15 #	99.5 ± 1.08 #	102 ± 2 #	98.8 ± 5.44 #	90 ± 1 #	253 ± 181 #	<b>120</b> 18-158
	Cr (mg/kg)	45.5 ± 0.4 #	44.9 ± 0.7 #	29.7 ± 0.66	28.6 ± 0.61	45.0 ± 0.5 #	44.9 ± 0.40 #	80 ± 2 *	47 ± 1 *#	<b>100</b> 102-136
	Cd (mg/kg)	3.25 ± 0.04 #	3.14 ± 0.08	2.21 ± 0.05	2.16 ± 0.09 #	2.2 ± 0.1 #	2.2 ± 0.1 #	3.4 ± 0.1 #	3.54 ± 0.35 #	<b>0.8</b> 0.4-3.9
Ni (mg/kg)	54.6 ± 0.6	55.7 ± 1.0	35.2 ± 0.72	35.4 ± 0.58	38.9 ± 0.6	37.8 ± 0.14	60 ± 1	62 ± 1.6	<b>30</b> 50-93	

**NOTES:**

\* indicate statistically significant differences among pairs (columns with some colour) (p < 0.05) – t-test

# indicate the absence of statistically significant differences inside the group of non-deposited and deposited SSA (p < 0.05) – ANOVA

<sup>a</sup> Danish EPA (Miljøstyrelsen)

<sup>b</sup> [5, 12, 14, 24, 28]

<sup>(1)</sup> samples used in the experiments aiming P-recovery (acid washing and ED process).

**Table 3.** Parameters measured at the beginning and at the end of the electrodialytic experiments.

Experiment	SSA suspension <sup>1</sup>						Mass loss (%)
	Voltage drop (V)		pH		Cond. (mS/cm)		
	Start	End	Start	End	Start	End	
F-3-0.19	5.1	4.2	2.25	1.61	22.2	11.7	41
D-3-0.19	6.8	4.4	2.04	1.84	32.4	13.2	40
F-2-0.08	6.7	4.3	2.63	1.68	9.6	22.5	43
D-2-0.08	5.7	4.7	2.47	1.55	14.1	32.8	41
F-2-0.19	5.0	4.5	2.11	1.28	15.9	32.6	42
D-2-0.19	4.8	4.7	2.80	1.24	26.7	33.3	36

<sup>1</sup> Central compartment in the 3 compartment cell and anode compartment in the 2 compartment cell

**Table 4.** MINTEQ calculation of major species of phosphate ions (%) for concentrations of elements in the the ash suspension liquid at the end of the treatments.

Experiments	F-3-0.19		D-3-0.19		F-2-0.08		D-2-0.08		F-2-0.19		D-2-0.19	
	Fe(II)	Fe(III)	Fe(II)	Fe(III)	Fe(II)	Fe(III)	Fe(II)	Fe(III)	Fe(II)	Fe(III)	Fe(II)	Fe(III)
<b>P</b>												
H <sub>2</sub> PO <sub>4</sub> <sup>-</sup>	22.6	21.2	28.8	27.2	27.6	27.6	22.8	22.6	14.6	14.2	13.7	13.3
H <sub>3</sub> PO <sub>4</sub>	55.3	51.4	42.3	39.8	57.4	57.1	67.2	66.6	73.7	71.6	76.5	74.2
AlHPO <sub>4</sub> <sup>+</sup>	4.62	4.28	7.73	7.28	4.50	4.44	2.29	2.23	1.73	1.68	1.39	1.34
Al <sub>2</sub> PO <sub>4</sub> <sup>+3</sup>	0.35	0.32	0.69	0.66	0.27	0.27	0.07	0.06	0.08	0.08	0.06	0.05
CaH <sub>2</sub> PO <sub>4</sub> <sup>+</sup>	4.48	4.17	5.77	5.43	5.96	5.92	3.17	5.92	3.71	3.62	2.84	2.76
FeH <sub>2</sub> PO <sub>4</sub> <sup>2+</sup>	12.6	12.9	14.6	10.9	4.31	3.09	4.48	3.75	6.17	7.30	5.55	7.05
FeHPO <sub>4</sub> <sup>+</sup>	-	-	-	8.68	-	-	-	1.64	-	1.49	-	1.33

**Table 1.** Mass of element removed to the anolyte *per* kg of ash at the end of the electrodialytic experiments (mg/kg).

<b>Elements</b>	<b>F-3-0.19</b>	<b>D-3-0.19</b>	<b>F-2-0.08</b>	<b>D-2-0.08</b>	<b>F-2-0.19</b>	<b>D-2-0.19</b>
P	33334.7	37879.6	102978.0	72153.4	124054.5	125420.4
Al	86.3	54.3	7957.8	5346.9	7148.3	9592.1
Cd	0.1	0.1	0.3	0.3	0.3	0.5
Cr	0.4	0.4	4.3	4.5	3.8	6.4
Cu	6.3	9.3	0.0	0.0	72.9	120.4
Fe	27.1	59.1	5246.1	6410.9	2867.8	4114.6
Ni	0.3	7.0	0.0	0.0	1.0	2.4
Pb	0.1	0.9	12.0	13.0	19.5	29.1
Zn	24.3	53.3	0.0	1.1	155.8	291.7