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Highlights

- Electrodialytic treatment of three Greenlandic fly ashes
- Cr (VI) is not the main Cr species after electrodialytic treatment
- Metal leaching from treated fly ash reduced to coal fly ash leaching levels

Electrodialytic treatment of Greenlandic municipal solid waste incineration fly ash

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7 Abstract

8 In Greenland, fly ash could contribute as a local resource in construction as a substitute for cement 9 in concrete or clay in bricks, if the toxicity of the ash is reduced. In this study, fly ash from three 10 different Greenlandic waste incinerators were collected and subjected to electrodialytic treatment 11 for removal of heavy metals with the aim of enabling reuse of the fly ashes. Seven electrodialytic experiments treating up to 2.5 kg of fly ash in a 10 L suspension were made. The heavy metal 12 removal was mostly dependent on the initial concentration in the fly ash. Heavy metal leaching was 13 14 examined before and after treatment and revealed overall a significant reduction in leaching of Cd, Cr, Cu, Pb and Zn; however, Cr and Pb leaching were above Danish guideline levels for reuse 15 purposes. Hg leaching was also reduced to below Danish guideline levels, although only 16 investigated for one fly ash. Hexavalent Cr was not the dominant speciation of Cr in the fly ashes. 17 Ettringite formed during electrodialytic treatment in the fly ash suspensions at pH above 12. The 18 19 total concentration of eligible components for reuse such as CaO, SiO₂ and Al₂O₃, increased during 20 the electrodialytic treatment.

21 Keywords: fly ash, electrokinetic remediation, hexavalent Cr, leaching, Arctic

22 1. Introduction

Incineration of municipal solid waste (MSW) is a well-established technology and accepted 23 24 treatment method for reducing the amount of MSW and at the same time recovering the energy of the waste. Residues after the incineration process consist of combustion products and unburned 25 waste and depending on the air pollution control, different reaction products. The main residue 26 27 fractions are bottom ash and fly ash. Typically, 10 - 30 kg fly ash is produced per ton of incinerated waste (Hjelmar et al., 2011). The fly ash is collected from the flue gas by electrostatic precipitators 28 (ESP) or bag filters. Due to the high content of organic and inorganic contaminants, fly ash is 29 30 classified as hazardous waste according to EU directive (91/689/EEC) on hazardous waste, whereas bottom ash does not classify as hazardous waste. In the US, fly ash is covered by the Resource 31 Conservation and Recovery Act (42 U.S.C. §6901 et seq. (1976)), and waste can be considered non-32 hazardous if leaching passes the Toxicity Characteristic Leaching Procedure (TCLP). Regardless of 33 these classifications, researchers are investigating treatment methods for fly ash and the 34 35 incorporation in different construction materials (Ferreira et al., 2003; Margallo et al., 2015; Quina et al., 2008; Zacco et al., 2014), to avoid the costly disposal as hazardous waste (Boesch et al., 36 2014). Generally, two approaches are considered when treating heavy metals in fly ash before 37 38 potential reuse: stabilisation or extraction of heavy metals. Stabilization methods include carbonation, thermal treatments or chemical stabilisations, such as stabilisation with phosphates, 39 40 chelating agents or ferrous compounds. In stabilisation, the material characteristics are only altered slightly, and the heavy metals are still present in the same quantity but are less available (Todorovic 41 and Ecke, 2006; Quina et al., 2008). Extraction techniques include different washing and chemical 42 43 extraction methods. Acid washing is efficient for removing heavy metals, but drastically alters the pH and material characteristics (Todorovic and Ecke, 2006; Quina et al., 2008). 44

One method that has shown potential for treating incineration residues is electrodialytic remediation 45 46 (EDR), where an electric current is applied to a contaminated material, and charged species, such as heavy metals are removed in the electric field. Most of the EDR research on incineration residues 47 has been made in laboratory scale, where suspending the residues in distilled water have been found 48 49 highly favorable for metal removal and experimental performance such as energy use and duration (Pedersen, 2002, Ottosen et al., 2006, Lima et al., 2012, Kirkelund et al., 2015, Viader et al., 2017, 50 51 Chen et al., 2018). A bench- and pilot scale electrodialytic (ED) set-up (Jensen et al., 2010) has also been developed for a continuous treatment method for Danish air pollution control (APC) residues. 52 The principle in ED treatment is shown in Fig. 1. The ED system consists of an ED stack with 53 multiple concentrate compartments (concentrate) and compartments containing the fly ash 54 suspension. Ion exchange membranes separate the compartments and the setup is designed to 55 remove anions and cations from the fly ash suspension to the concentrate by the applied current. 56 57 The early aim of EDR in laboratory scale was to remove the largest possible fraction of heavy metals, which require a significant pH reduction of the material, acquired primarily by water-58 59 splitting at the anion exchange membrane (Ottosen et al. 2000) or assisting agents. This 60 acidification implies long treatment times. Removals up to 90% Cd, 85% Cu, 60 % Zn, 45 % Cr 61 and 20 % Pb has been achieved after 70 days removal with ammonium citrate as assisting agent for MSWI fly ash (Pedersen et al., 2005). However, both the acidic pH and the use of assisting agents 62 alters the treated material's matrix significantly. This is seen by dissolution of up to 78 % fly ash 63 64 when acidified (Ottosen et al., 2006), adding new mineral phases to the fly ash and altering the morphology (Pedersen and Gardner, 2003) or even increasing the mass of treated fly ash by up to 65 12 % when using assisting agents (Pedersen et al., 2005). The later ED experiments in bench - and 66 pilot scale focused on the reduction of the metal and salt leaching to reduce environmental 67 hazardousness while leaving the non-mobile fractions in the ED treated material. Thus, larger 68

amounts of material can be treated and shorter retention times kept. Pilot scale experiments with 69 both Danish fly ash and APC residue showed reduced leaching of chloride and most metals (Jensen 70 et al., 2015; Kirkelund et al., 2010). One element that showed significantly increased leaching in the 71 studies with the Danish residues was Cr. The increased Cr leaching seen in these studies could be 72 73 due to changes in Cr speciation from Cr (III) to the more mobile and toxic Cr (VI) during the electrodialytic treatment for the Danish fly ash and APC residues. The Cr behaviour during ED is 74 not well understood, however, the solubility and speciation of Cr in treated ED materials are 75 important for reuse options. An important finding by (Jensen et al., 2015) was that fly ash without 76 77 flue gas cleaning products appear more suitable for ED treatment than if flue gas cleaning products are mixed into the fly ash. 78

79 Solid waste has typically been dumped at uncontrolled landfills in the Arctic (Poland et al., 2003). Use of uncontrolled landfills was also the practice in Greenland before first waste incinerator started 80 operating the 1990ies, primarily to limit the amount of waste for disposal and hygienize the waste. 81 82 There are six Greenlandic MSWI plants in the major towns in Greenland. Greenland has a total 83 population of 56,000 people. The Greenlandic incinerators are smaller and simpler than traditional European waste incinerators and have a capacity between 2,000 – 10,000 t annually (Eisted and 84 85 Christensen, 2011) and represent a waste management option for smaller isolated communities. The grate furnace incinerators in Nuuk, Sisimiut and Qaqortoq have flue gas treatment only in the form 86 of electrostatic precipitators (ESP) and the grate furnace incinerators in Maniitsoq, Ilulissat and 87 Aasiaat have cyclones followed by electrostatic precipitators. This equipment is capable of 88 89 removing only larger fly ash particles (Buonanno and Morawska, 2015) whereas bagfilters would 90 remove down to ultrafine particles (Jones and Harrison, 2016) and there is no acid gas cleaning in any of the Greenlandic incinerators. All incinerators have heat boilers for energy recovery. The 91 resulting residues from the incinerators are bottom ash, boiler ash and fly ash. The boiler and 92

bottom ashes are collected and disposed of or used as cover material at the uncontrolled landfills.
The hazardous fly ash is stored in big bags at the uncontrolled landfills before being shipped for
deposition at hazardous waste disposal sites in Europe. This shipment and disposal is a considerable
expense for the Greenlandic municipalities. Safe handling of hazardous waste is of interest in the
Arctic due to the vulnerable environment, which can suffer severe consequences due to
contamination from waste (Gilbreath and Krass, 2006; Poland et al., 2003; Weichenthal et al., 2015;
Weiser and Powell, 2011).

Additionally, the possible resources in the fly ash are lost for later utilisation by disposal. 100 101 Construction materials are widely imported to Arctic regions, including Greenland. Even though local secondary resources in Greenland are scarce, the use of MSWI fly ash is limited to the 102 hazardousness of the ash. Therefore, there is a need to remove or stabilise the contaminants in the 103 ash. All previous EDR experiments are made with fly ash from incinerators with advanced flue gas 104 cleaning systems and the metal removal is dependent on the ash type (Lima et al., 2012). These 105 106 results cannot be transferred to the Greenlandic fly ashes directly, since these incineration plants are smaller and less advanced. Thus, it is necessary to study the Greenlandic fly ashes separately to 107 evaluate the treatment potential. The aim of this study was therefore to investigate the possibility to 108 109 upgrade Greenlandic MSWI fly ashes by electrodialytic treatment to obtain a stable residue that can be reused in construction by 1) investigating leaching levels of Greenlandic fly ash before and after 110 ED treatment 2) investigating the uniformity of fly ashes from three Greenlandic plants with 111 similar fly ash collection systems after electrodialytic treatment and 3) investigating the influence of 112 current density and liquid to solid ratio on the environmental performance, including Cr (VI), which 113 114 is previously unexamined.

116 **2. Materials and methods**

117 **2.1.** Experimental fly ashes

ESP fly ashes (without acid gas cleaning products) were collected from big bags at the temporary 118 119 storages at random days from the incinerators in Nuuk, Sisimiut and Qaqortoq, which have annual incineration capacities of 10,000; 4,000 and 3,000 tons respectively (Eisted and Christensen, 2011). 120 121 No information about the specific type of waste incinerated in the period of sampling exists. The 122 collected fly ash samples were stored in sealed boxes at room temperature before the experimental work. The fly ashes were sieved through a 1 mm sieve before the electrodialytic experiments not to 123 block the ED stack. For the Sisimiut and Qaqortoq fly ashes the amount passing the 1 mm sieve was 124 100 % and for the Nuuk fly ash, 92 % passed the 1 mm sieve. 125

126 **2.2. Analytical procedures**

Total concentrations and leaching of heavy metals (Cd, Cr, Cu, Pb, Zn) in the fly ash were 127 measured by induced coupled plasma- optical emission spectrometry (ICP-OES). Five digestions of 128 129 each fly ash were made after the Danish Standard DS259 where 1 g of fly ash and 20 mL 7.3 M 130 HNO₃ were heated at 200 kPa (120°C) for 30 min. The liquid was after that separated by vacuum filtration through a 45 µm filter and diluted to 100 mL. Leaching experiments were made according 131 132 to Danish standard DS/EN 12457-3 part 1, three for each fly ash. The liquid to solid (L/S) ratio was 2, mixing 40 g fly ash and 80 mL of distilled water. The suspension was shaken for 6 hours on an 133 134 end-over shaker before vacuum filtration through a 45 µm filter. Selected digested and leached samples were measured for Hg by atomic absorption spectrometry (AAS) with a hybrid system by 135 an external laboratory. Water content was measured by weight loss at 105°C for 24 h. Loss on 136 ignition was measured at 550°C for one hour. The pH was measured in 1 M KCl at a liquid-to-solid 137 ratio (L/S) of 5 and after 1 hour of agitation, pH was measured by a Radiometer Analytical pH 138

electrode. The amount of water-soluble fly ash was estimated as mass reduction when mixing 1 g 139 140 fly ash with 20 mL of distilled water which was agitated for 24 hours. pH-dependent batch extraction experiments were made with HNO3 and as a reference distilled water was used. The 141 extractions were made by mixing 10.0 g fly ash with 50 mL HNO₃ in concentrations of 0.01 M -142 143 0.7 M in 100 mL acid rinsed plastic bottles. For the Qagortoq sample, NaOH in concentrations of 0.02 - 0.05 M was also added to increase the pH. All the extractions were made in triplicates and 144 shaken for 72 hours on a horizontal shaker operating at 250 rpm to keep the fly ash in suspension. 145 After that the samples settled for 15 minutes before pH was measured. The extractants were filtrated 146 through a 45 μ m filter and acidified if pH > 2 by addition of concentrated HNO₃ before 147 measurements of metals by ICP-OES. Extractions in triplicate on solid fly ash samples for Cr (VI) 148 total analysis were made according to the US EPA 3060A method. Cr (VI) in the solutions was 149 measured by colourimetrically UV-VIS spectrophotometry on a Spectroquant Nova 60. 150

151 Major oxide composition was estimated from semi-quantitative analysis by X-ray fluorescence 152 (XRF) on powder samples by an external laboratory. Ash mineralogy was studied by X-ray powder 153 diffraction (XRD), for identification of major crystalline phases. The instrument was a PANalytical 154 X'Pert Pro operating at 45 mA and 40 kV applying Cu K α radiation with a 2 Θ X'Celerator detector. 155 The samples were scanned in the range of 4-70 2 Θ within 2.5 hours. The diffractograms were 156 interpreted by using the ICDD PDF-4 database for minerals. XRF and XRD measurements were 157 made on the raw fly ash samples and selected ED treated samples.

158 2.3. Electrodialytic experiments

Seven electrodialytic experiments were made with a bench scale stack set-up and the operational
conditions varied. The experimental conditions shown in Table 1 were chosen to find the optimal
applied current density and liquid to solid ratio (L/S) on different samples of fly ashes. In a previous

study with APC residues, L/S between 10-17 were used in a smaller bench scale stack set-up (Jensen et al., 2010). Lower L/S ratios were chosen in this study to increase the amount of material to be treated and hence the capacity of the treatment method. Compared to previous laboratory experiments, the used power supply could maintain a much higher current (max. 10 A) compared to the power supply used for the laboratory cell (max. 0.25 A) (Kirkelund et al., 2015), thus higher applied currents were tested in this study to investigate if a shorter experimental duration was possible with higher current densities and total applied charge.

169

The ED stack consisted of 5 cell pairs (530 cm² active surface membrane area per cell) and Neosepta cation and anion exchange membranes were used. The spacers were 0.6 mm and 5 mm thick for the concentrate and fly ash suspension compartments respectively. The spacers used for the fly ash suspension were thicker than the concentrate spacers to avoid clogging due to the high solids content in the fly ash suspension. In Fig. 1 it is seen how the ion exchange membranes and flow spacers were mounted in the ED stack.

176 There were electrode compartments at each end of the ED stack. The electrodes were titanium 177 plates covered by a layer of metal oxides. A constant current of either 10 or 5 A was applied to the ED stack, resulting in current densities of 9.4 or 4.7 mA/cm², by 530 cm² active surface membrane 178 area. The electrode compartments shared electrode solution, which was circulating. The pumps 179 180 circulating the concentrate, fly ash suspension and electrode solution were for experiments N-1 and N-2 centrifugal pumps. For the remaining experiments, the pump for the fly ash suspension was a 181 peristaltic pump PSF3 from Ragazzini. The concentrate and electrolyte solutions were 5 L each of 182 distilled water adjusted to pH 1 with HNO₃ to avoid precipitation of metals in the pumps, spacers 183 and electrodes. The fly ash suspensions were made by mixing 1 - 2.5 kg fly ash with 10 L distilled 184

water. Electrical conductivity and pH in the fly ash suspension were measured every 30 minutes of
the experiments as well as the voltage over the ED stack, pH and electrical conductivity of the
concentrate and electrode solution. Compared to the laboratory electrodialytic experimental cell
(Kirkelund et al., 2015), the removal of both anions and cations were to the same solution,
concentrate and/or electrode solution.

After the experiments, the fly ash particles were separated from the liquid by filtering the suspension through 45 µm filters. The fly ash was dried at 40°C before digestions for measuring the total heavy metal content, pH, leaching experiments and also XRF and XRD analysis for selected samples. The volumes of the filtered liquid solution, concentrate and electrode solution were noted and samples were taken for metal measurements on ICP-OES. The ED stack was taken apart and thoroughly rinsed with water between each experiment.

The metal removal in the electrodialytic experiments is defined as the amount of metals found in the concentrate and electrode solutions compared to the initial amount of metal in the fly ash and was calculated for each metal in each experiment:

199 Removal (%) = ((CL_{concentrate}×V_{Concentrate}+ CL_{electrode} solution×V_{Concentrate} solution)/C_{fly ash} × M_{fly ash}) × 100 200 %

Where CL (mg/L) is the measured concentration in the concentrate and electrode solution, V (L) the volume, C (mg/kg) the concentration in the fly ash and M (kg) the initial mass of fly ash in the experiment.

204 **3. Results and discussion**

205 **3.1. Fly ash characterisation**

206 The characterisation showed significant differences between the three fly ashes even if the incineration plants have the same flue gas treatment systems, see Table 2. The Nuuk and Sisimiut 207 fly ashes have the characteristic alkaline pH of MSWI fly ash, but the Qaqortoq fly ash had a 208 neutral pH. This lower pH of the latter fly ash is not unusual (Hjelmar et al., 2011) and could be due 209 210 to the technical performance of the fly ash collection and cooling of the flue gas in the incineration plant. The composition of the fuel and the combustion process are important factors for 211 determining the heavy metal content in the fly ash, especially Cl influences the amount of Pb in the 212 fly ash (Pedersen et al., 2009). Regardless, the total concentrations of Cd, Cr, Cu, Pb and Zn were in 213 the same orders of magnitude for the three fly ashes. The fraction of Cr determined as Cr (VI) was 214 low compared to the total Cr concentration in all three ashes and Cr (VI) was not the dominant 215 speciation of Cr in the fly ashes before the ED. These results show that the ESP ash from the small-216 scale Greenlandic incinerators was within the typical ranges of fly ash from larger incineration 217 plants (Lam et al., 2010). Fig. 2 shows the pH-dependent desorption of the heavy metals. It can be 218 seen that the pH-dependent desorption followed a similar pattern for the metals in the three different 219 ashes. Cd, Cu and Zn were desorbed at acidic pH mainly, whereas Cr and Pb to some extent also 220 were desorbed at alkaline pH. Zn desorption from the Nuuk ash started below pH 4 and was 221 222 desorbed to a lower degree than for the Sisimiut and Qaqortoq ashes. Generally, the maximum desorption of the metals (~100 %) was seen at pH 2, although the desorption degree from the Nuuk 223 ash was between 70-80 % for the metals. The different fractions that fly ash typically consist of are 224 225 soluble in different pH ranges; oxides between 2 < pH < 6, carbonates 6 < pH < 9 and hydroxides 10 < pH < 13 (Flyvbjerg and Hjelmar, 1997). The release of metals in these pH ranges could 226 227 indicate preferable speciation of the metals towards these phases. Compared to the experimental results in Fig. 2, the highest desorption was seen in the pH range from 0 to 7, indicating dominating 228 oxide speciation, and for Cd and Zn, also carbonates, as desorption was seen around pH 7. A 229

different trend was observed for Cr, where up to 40 % was desorbed above pH above 10. This
indicates very different speciation of Cr in the fly ashes, possibly as hydroxides. Lima et al. (2008)
suggested that at pH above 12 in fly ash, Cr desorbed as Cr (VI).

233 **3.2. Electrodialytic experiments**

In Fig. 3, pH and electrical conductivity in the fly ash suspension, as well as the voltage drop overthe ED stack, are shown for the experiments.

The voltage drop was not directly linked to the electrical conductivity in the fly ash suspension, so 236 237 the high resistance resulting in the maximum voltage of the power supply in experiments N-1 and 238 N-2 could result from either resistance over the membranes or a low conductivity of the concentrate. The applied current density could not be kept constant in these experiments as the 239 240 maximum voltage of 32 V for the power supply was reached after about 1 hour at a current density of 9.4 mA/cm2 and around 5 hours at current density 4.7 mA/cm2. Problems with clogging in the 241 system were seen with L/S 5 and 4. The electrical conductivity in the fly ash suspension decreased 242 with time due to the continuous removal of ions in the stack system. pH in the fly ash suspensions 243 decreased only slightly throughout the experiments, which was expected due to the buffering 244 245 capacity. The ED system acidified harbour sediment suspensions at much lower current densities (< 0.12 mA/cm2) after 8-16 hours (Pedersen et al., 2015), but the buffering capacity of fly ashes is 246 much higher than that of harbour sediments. 247

248 **3.3. Metal removal and distribution**

The distribution of heavy metals in the ED stack compartments (electrolyte, concentrate and suspension) after the electrodialytic experiments is shown in Fig. 4, together with the removal. The suspension amount represents the amount of metal that is found in the liquid after the fly ash suspension was filtered. The removed amounts increased for all metals, except Cr, with decreasing 253 L/S ratio. The removal was also influenced by the pH in the fly ash solution. The highest removal for Cd, Cu and Zn was seen in the Sisimiut and Qagortog fly ash suspension, which had lower pH 254 than the Nuuk fly ash suspension. The highest removals were 15 % Cd and 49 % Cu in the Q-1 255 experiment and 7.4 % and 10.1 % Pb in N-1 and N-2. All other removal rates were below 6 %. 256 257 Previous work has shown removals of up to 55 % Cd, 4.5 % Cr, 90 % Cu, 66 % Pb and 55 % Zn from fly ash in water suspensions, which reached final pH 3.9 - 8.2 in electrodialytic experiments 258 (Ottosen et al., 2006, Kirkelund et al., 2015). This pH was only achieved in the Q-1 experiment, 259 where the removals were 15 % Cd, 49 % Cu, 2 % Cr, 3 % Pb and 5 %. The removals do not take 260 into account any amount of metal that has electrodeposited on the cathode, which readily occurs for 261 Cu and Pb (Ottosen et al., 2003, Ottosen et al., 2006). The efficiency of electrodeposition of metal 262 cations is dependent on several factors such as electrode material, current density, electrode surface 263 area and the electrolyte solution matrix (Chen, 2004) and could underestimate the removals in this 264 study. Lima et al. (2010) showed that ash type, duration of the electrodialytic experiment and 265 dissolution of fly ash during electrodialytic remediation to be the most important factors for metal 266 removal. The duration of the experiments was short, however, the applied current higher than in 267 previous experiments. 268

269 The amount of metal found in the filtered suspension liquid indicates released metal compounds that are uncharged and which cannot electromigrate and stay in the suspension. The highest amount 270 found in the suspension was for Cr in all experiments except N-1 and N-2 and Pb in the N-1 and N-271 2 experiments. Pb forms stable complexes with Cl and Pedersen (2002) predicted that in a fly ash 272 solution with a high concentration of Cl, 35 % of Pb would be present as uncharged PbCl₂. Also at 273 274 the high pH in the Nuuk experiments, Pb(OH)₂ could be a dominant species. Thus, a lower pH favours a larger fraction of charged Pb species, which is removed in the electric field as seen in all 275 276 Sisimiut and the Q-1 experiments.

The amount of Cr in the suspension liquid was significant, regardless of pH in the solution and 277 operational conditions. Thus, the speciation and behaviour of Cr in the ED stack differ significantly 278 to the other investigated metals. Hexavalent Cr would be on anionic form and electromigrate 279 towards the anode and trivalent Cr on cationic form and electromigrate towards the cathode. 280 281 However, with the combined electrode solution and the concentrate, it is not possible in this experimental setup to determine the charge of the removed Cr. There also seems to be a dependence 282 on the type of fly ash, as a similar amount of Cr in the suspension liquid has not been seen for 283 Danish fly ash nor APC residue (Kirkelund et al., 2015). Cr(OH)3 could be the dominant species in 284 the solution in the pH range in the experiments (pH 7-12) (Ščančar and Milačič, 2014). Although 285 the Cr was found in the suspension liquid, it is released from the fly ash particles, and this is 286 consistent with results from the pH-dependent desorption (Fig. 2), which showed a high release of 287 Cr at alkaline pH. 288

289 3.4. Heavy metal content and leaching

The final heavy metal concentrations in the ash and the leaching from the fly ashes are seen in 290 291 Table 3. Total metal concentrations in the fly ashes typically increased with ED treatment. This was due to the substantial dissolution of fly ash and regardless of heavy metal removal. Jensen et al. 292 (2015) suggested that the solubility of the fly ash, especially the amount of soluble salts, is 293 294 determining the final metal concentrations, as the metal species are less soluble and the metals will thus be concentrated in the less soluble parts remaining after treatment. The solubility of the ashes 295 was in the order Qagortoq > Nuuk > Sisimiut, and direct relationship between the solubility and 296 297 increase in total concentrations by the ED treatment was observed for most metals. The final concentrations were less influenced by the differences in L/S ratio, current density or metal removal 298 during the ED treatment. 299

Generally, the heavy metal leaching decreased during ED treatment (Table 3). Table 3 also shows 300 the Danish limiting values C3 that represent the maximum allowed leaching values for reuse of 301 waste materials not classified as hazardous for geotechnical purposes in Denmark (Miljøstyrelsen, 302 2015) and leaching concentrations of coal fly ash (CFA), which is widely used in construction and 303 304 as supplementary cementitious material. Comparing the leached values to the Danish limiting value C3 it is seen that none of the ED treated samples comply with the values for all metals. Pb and Cr 305 exhibited the highest exceeded leaching compared to C3 and only Pb leaching in experiment N-1 306 and Q-1 was below the C3 value. Compared to CFA, the Pb leaching in the electrodialytically 307 treated fly ashes was lower, except for experiment N-2. The leaching of Cd was reduced to below 308 the C3 value for experiments N-1, N-2, S-1 and S-2. Hg was measured in selected samples only and 309 the leaching decreased to below the C3 value in experiment S-1. In all experiments, the leaching of 310 Cu and Zn was below the C3 value and similar to leaching from CFA. The Cu leaching was also 311 312 below this value in the raw fly ashes, whereas Zn leaching was below the C3 value for the raw Sisimiut fly ash, but not for the Nuuk and Qaqortoq raw ashes. The reduction in leaching is linked 313 to the metal removal and also to the decrease in pH of the ashes during electrodialytic treatment, 314 where metal leaching is lowest in the more neutral pH range. 315

316 Cr leaching decreased due to the ED treatment for all Sisimiut experiments and experiment N-2, contrarily to what has been seen for Danish APC residues and fly ash where the Cr leaching always 317 increased (Jensen et al., 2015; Kirkelund et al., 2010; Kirkelund et al., 2015). The total Cr 318 concentrations in the solid fly ash increased for all fly ashes except S-1 and S-4 and the hexavalent 319 320 Cr concentrations in the solid fly ash were non-detectable or low compared to the total Cr 321 concentration. This indicates that if present, the hexavalent Cr is removed but not formed during ED treatment. For most fly ash samples, the Cr (VI) measurement was below the detection limit of 2 322 mg/kg. Based on the L/S ratio for the analysis this corresponds to a leached Cr value of 1 mg/l, and 323

when comparing this value to the actual leached Cr values, it suggests that Cr (VI) might play a
significant role in the leaching behaviour. The leached Cr concentration in experiment S-3 and S-4
is at a similar level as CFA. These results are in contrast to findings from Danish fly ash and APC
residues, where the Cr leaching rarely exceeds C3 or CFA leaching values after electrodialytic
treatment, but Pb and Zn do (Jensen et al., 2015; Kirkelund et al., 2015; Kirkelund et al., 2010).
Thus, leaching seems to depend on the type of flue gas treatment and possibly also the type of fuel
for Cr behaviour.

There is a need to find a suitable method for treated MSWI fly ashes to avoid expensive disposal of 331 332 this waste material. According to legislation, MSWI fly ash is classified as hazardous waste due to the high content of inorganic and organic contaminants. Most treatment techniques have been 333 applied to study the stabilisation or reduction of the inorganic contaminants. The main challenge for 334 the valorization of MSWI fly ash is that there does not exit end-of-waste (EoW) criteria or other 335 regulatory limits for leaching or total metal concentrations that apply to MSWI fly ash due to the 336 337 classification as hazardous waste. Even if the EoW does not exist, it is of interest to compare the 338 leaching concentrations from the ED treated fly ashes in this work to other treatment methods to evaluate the applicability of ED. Leaching concentrations after different treatments are shown in 339 340 Fig. 5, together with guideline limits for leaching set by the TCPL leaching test, the Danish Category 3 and Danish limits for disposal of inert waste. 341

The treatment methods are divided into washing with different liquids, thermal methods, electrokinetic and as combinations, as several treatment methods are often used together. Several studies apply treatment methods before a valorisation test in construction material; however, only a few of these studies give information about leaching of the treated fly ash itself. They rather present the leaching results from the construction material into which the treated fly ash was incorporated, and these leaching results present a dilution of the leaching of the treated fly ash. The leaching after

different treatments of Cd, Cr, Cu, Pb and Zn can be seen to vary up to 5 orders of magnitude, 348 which implies the difficult nature of the fly ashes, as shown in Fig. 5. Also, some leaching tests 349 were made with the TCPL test using acetic acid instead of distilled water according to the DS/EN 350 12457-3 leaching test commonly applied in Europe. The leaching results with the TCPL tests are 351 352 thus naturally higher than with the DS/EN 12457-3 test. The leaching results of the ED treated fly ash in this work were within the range of other MSWI fly ashes treated with different methods. 353 Very few of the methods from the literature make the leaching stand out as particularly low or 354 below the limits they are compared with for all metals studied. As the heavy metal leaching is 355 highly pH dependent and each treatment method result in different pH of the MSWI fly ash, the 356 leaching is highly dependent on final pH. Although, a combination of treatments seems to be the 357 most viable option, commonly for all methods is a significant decrease of the initial leaching of the 358 fly ashes (Chen et al., 2017; Liao et al., 2014; Liu et al., 2009; Lundtorp et al., 2002) which is 359 beneficial for valorization. As an oxyanion Cr behaves differently from other metals and even 360 increased leaching of Cr has been observed when fly ash is treated by thermal treatment or water 361 washing (Liu et al., 2009; Lundtorp et al., 2002). This increased leaching can be due to oxidation of 362 trivalent Cr to hexavalent Cr at high temperature (Wang et al. 2001) and an increase in the soluble 363 fraction of Cr as calcium chromate when in reaction with CaO and CaCl₂ during thermal treatment 364 (Kirk et al., 2002). When ash is in contact with water, hydroxide ions are formed by hydrolysis of 365 metal oxides, and this increases the pH, which favours Cr leaching (Nordmark and Lagerkvist, 366 367 2018). This hydrolysis continues in the washed ash even after the washing process has ended, and is the reason for increased Cr leaching in water/acid treated ash compared to raw ash (Nordmark and 368 Lagerkvist, 2018). It is, therefore, necessary to control pH and redox conditions in the fly ash to 369 370 reduce the Cr leaching.

371 **3.5 Matrix changes**

The XRD diffractograms are seen in Fig. 6 from where the major crystalline phases in the fly ashes 372 373 before and after ED treatment are specified. The major components in the fly ashes before and after ED treatment are shown in Table 4 and compared to major components in cement, clay and coal fly 374 ash (CFA). The untreated fly ashes consisted mainly of Ca and Cl, and Na, K and Al to a lower 375 376 degree (Table 4). The main mineral phases of the fly ashes were also linked to these elements and classified as Ca-containing minerals such as anhydrite (CaOH) and carbonate (CaCO₃) and Cl-377 378 containing minerals such as sylvite (KCl) and halite (NaCl). Formation of ettringite (Ca₆Al₂(SO₄)₃·26H₂O), which is an indication of ageing of the ash (Steenari et al., 1999) were 379 seen in both the Nuuk ED experiments and the S-2 experiment, but not in the other fly ash samples 380 after ED treatment. The formation of gypsum (CaSO4·H2O) was significant for the Sisimiut and 381 Qagortoq fly ash during ED treatment as opposed to the Nuuk fly ash. Formation of ettringite rather 382 than gypsum is dependent on pH and has found to be favoured at pH between 11 and 12.5 383 384 (Chrysochoou and Dermatas, 2006), which also coincides with the pH of the Nuuk fly ash suspensions as seen in Fig. 3. The Zn-containing mineral simonkolleite (Zn₅(OH)₈Cl₂·H₂O) was 385 identified for the Qagortoq fly ash after ED treatment, which corresponds well to the high Zn 386 concentration after the EDR treatment. During ED treatment soluble minerals such as the Cl-387 minerals dissolved and could be removed in the electric field. Na, K and Cl were removed from the 388 fly ashes, but to a greater extent for the Nuuk ash than Sisimiut and Qaqortoq, which could be due 389 390 to the longer duration time or the low L/S ratio. This is also shown in the XRD diffractograms: 391 halite was present in the Sisimiut and Qaqortoq ED treated fly ashes as opposed to the Nuuk ED treated ash. Thus, increasing the treatment time increased the Cl removal, which was also seen in 392 another study by (Kirkelund et al., 2015) in laboratory scale EDR experiments. 393

CaO, SiO₂ and Al₂O₃ are major components that are desirable for the valorisation of fly ash as
substitution of primary resources in concrete, glass-ceramics, ceramic products and glass materials

(Chu et al., 2013; Haiying et al., 2011; Rémond et al., 2002). The total amount of CaO, SiO₂ and 396 397 Al₂O₃ was 53%, 33% and 30% for the Nuuk, Sisimiut and Qaqortoq fly ashes before ED treatment. After ED treatment this amount had increased to 58 %, 48 % and 33 %, mainly due to the 398 dissolution of soluble salts, here seen as the depletion of K₂O, Na₂O and Cl in the ED treated fly 399 400 ash samples. The fly ashes did not directly resemble cement or clay in composition but was more similar to coal fly ash (CFA). CFA is widely used as a substitute for cement in concrete, however, 401 402 fly ash from waste incineration is not at present, but this could change in the future due to higher disposal costs and more focus on secondary resources if the heavy metal leaching can be reduced 403 (Zacco et al., 2014). 404

The pH-dependent leaching of the Greenlandic fly ashes could suggest that reaching a lower pH of 405 the fly ashes (around pH 6) might be better for reduced Cr leaching as well as the other studied 406 metals. However, at such pH reduction, the material characteristics of the ED treated fly ash is 407 expected to be different due to a larger dissolution of minerals, and the incorporation in alkaline 408 409 construction materials could pose challenges. By the current and previous results, it is recommended to test each treated fly ash case by case when valorised and also do a long-term 410 evaluation of metal leaching and the environmental impact of the after-life, when the construction 411 412 material with the incorporated fly ash becomes waste again.

413 **4. Conclusion**

The composition of Greenlandic fly ash varied among the three investigated incinerators, even though the flue gas treatment systems were similar. The heavy metal removal was more dependent on the fly ash and initial heavy metal content rather than the experimental conditions such as current density, duration and L/S ratio. Cr was not removed from the liquid phase of the fly ash suspension to the concentrate to the same degree as the other metals. This could be due to the speciation and

419	formation of uncharged species. Due to the dissolution of fly ash during ED treatment, there is an
420	increase in both heavy metal concentrations and major elements and minerals content. Hexavalent
421	Cr is not the dominant speciation of Cr in the raw fly ashes and is not formed during electrodialytic
422	treatment. Leaching limit levels for use in geotechnical purposes were exceeded in the raw ashes for
423	Cd, Cu, Hg, Pb and Zn. After ED treatment these leaching limits were met for Cu, Hg and Zn.
424	Leaching of Cd, Cr and Pb also decreased after ED treatment, but the regulatory leaching limits
425	could not be reached. Although the leaching of the ED treated fly ashes was similar to leaching
426	from certified coal fly ash used in construction materials. There is a valorisation potential of the ED
427	treated fly ashes in Greenland for use in construction materials, and this should be investigated
428	further with material tests, including how heavy metals are incorporated.

429

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Fig. 1 ED stack and one cell pair set-up



 \bigcirc Nuuk \diamondsuit Sisimiut \triangle Qaqortoq

Fig. 2 pH dependent extraction of Cd, Cr, Cu, Pb and Zn from the fly ashes



 \bigcirc N-1 \bigcirc N-2 \diamondsuit S-1 \diamondsuit S-2 \blacklozenge S-3 \blacklozenge S-4 \blacktriangle Q-1

Fig. 3 pH and electrical conductivity in the fly ash suspensions during the ED experiments and voltage over the stack



Fig. 4 Amounts (mg) of metals dissolved in the different compartments of the stack after ED treatment and total removal (%)



Fig. 5: Leaching concentrations (mg/kg) from fly ash after different treatments and regulatory limits. * - TCLP-leaching method, ¹EPA 2004, ²Miljøstyrelsen 2015, ³Miljøstyrelsen 2011, ⁴Lui et al. 2009, ⁵Belmonte et al. 2017, ⁶Lundtorp et al. 2002. ⁷Chen et al. 2017, ⁸Jensen et al. 2015, ⁹Liao et al. 2014, ¹⁰Aubert et al., 2006



Fig. 6 XRD diffractograms with main mineral phases before and after ED treatment. A-anhydrite, C-carbonate, E-Ettringite, G-gypsum, H-halite, Si-Simonkolleite, S-sylvite.

Table 1 Experimental conditions for the electrodialytic experiments

Experiment	Fly ash	L/S ratio	Duration	Applied current	Current density
			(hours)	(A)	(mA/cm ²)
N-1	Nuuk	10	10	5	4.7
N-2	Nuuk	10	10	10	9.4
S-1	Sisimiut	10	5	5	4.7
S-2	Sisimiut	6.7	5	5	4.7
S-3	Sisimiut	5	5	5	4.7
S-4	Sisimiut	4	5	5	4.7
Q-1	Qaqortoq	6.7	5	5	4.7

Parameter	Nuuk	Sisimiut	Qaqortoq	ESP fly ash ¹
рН	12.5	12.5	7.2	-
EC (mS/cm ²)	79	81	43	-
Water content (%)	0.6	0.6	2.5	-
Water solubility (%)	34	23	47	-
Cd (mg/kg)	314	447	210	5-2,211
Cr total (mg/kg)	165	177	115	21-1,901
Cr (VI) (mg/kg)	3.8	<2	2.2	-
Cu (mg/kg)	718	1,540	713	187-2,831
Hg (mg/kg)	0.4	8.8	-	0.8-73
Pb (mg/kg)	1,330	5,020	3,370	200-19,000
Zn (mg/kg)	13,200	38,300	36,600	2,800-150,000

Table 2 Characteristics of experimental fly ashes. ¹ Lam et al. 2010

		Nuuk	N-1	N-2	Sisimiut	S-1	S-2	S-3	S-4	Qaqortoq	Q-1	C31	CFA ²
	Cd (mg/kg)	314	462	407	447	362	474	827	682	210	272	>0.5	1.5
	Cr (mg/kg)	166	457	401	177	156	228	195	160	115	237	>500	48
tent	Cr (VI) (mg/kg)	3.8	<2	<2	<2	<2	2.7	4.9	<2	2.2	2.5	>20	-
l con	Cu (mg/kg)	718	1,310	1,140	1,540	1,530	2,080	2,160	1,770	713	1,680	>500	32
Tota	Hg (mg/kg)	0.41	0.14	-	8.8	15	-	-	-	-	-	>1	-
	Pb (mg/kg)	1,330	2,650	2,170	5,030	5,720	8,060	6,760	5,420	3,370	6,920	>40	30
	Zn (mg/kg)	13,200	22,800	21,000	38,300	39,700	53,000	59,700	42,800	36,600	78,100	>500	140
	рН	12.3	10.9	12.6	12.4	9.9	10.0	9.5	9.0	10.2	8.0	-	12.9
	Cd (µg/l)	35	<20	<20	1460	<20	32	580	3612	3900	62	40	62
gu	Cr (mg/l)	5.4	5.6	1.7	11.5	3.7	3.7	1.2	1.1	1.6	1.9	0.5	1.1
achii	Cu(µg/l)	300	61	41	74	65	76	80	80	60	<20	2000	75
Le	Hg (µg/l)	1.6	-	-	1.2	0.6	-	-	-	-	-	1	-
	Pb (mg/l)	37.3	0.08	4.4	0.5	0.36	0.42	0.42	0.54	2.1	< 0.02	0.1	0.9
	Zn (mg/l)	5.5	0.4	1.0	1.1	0.33	0.18	0.26	0.32	3.6	0.02	1.5	0.4

Table 3 Leaching and total concentrations in the fly ash samples before and after electrodialytic treatment. ¹Miljøstyrelsen 2015, ²Jensen et al. 2015.

	Nuuk	N-1	Sisimiut	S-2	S-3 ¹	Qaqortoq	Q-1	Cement ²	Clay ^{3,4}	CFA ⁵
CaO (%)	42.0	44.8	25.2	30.8	26.3	15.4	18.2	60.6-66.3	0.4-2.5	0.5-54.1
SiO ₂ (%)	6.2	8.1	4.9	10.5	9.3	2.8	9.8	18.7-22.0	61.5-70.6	14.8-56.7
Na ₂ O (%)	9.6	0.1	16.2	3.1	6.4	14.8	<0.7	0.11-1.20 (Na ₂ O	0.5	0.04-1.23
K ₂ O (%)	6.5	0.2	8.9	2.0	3.7	12.0	2.4	equivalent)	2.7-3.1	0.2-4.89
$Al_2O_3(\%)$	4.9	5.3	3.2	6.6	5.3	1.2	5.1	4.7-6.3	14.8-15.8	3.4-29.2
$Fe_2O_3(\%)$	0.8	1.3	1.3	1.4	1.6	0.7	1.1	1.6-4.4	4.5-6.1	0.86-11.8
MgO (%)	1.7	1.6	0.6	2.0	1.9	0.1	1.2	0.7-4.2	1.3-2.2	0.2-9.1
MnO (%)	0.1	0.1	0.04	0.05	0.07	0.1	0.05	-	0.8	0.02-0.21
$P_2O_5(\%)$	1.0	1.7	0.6	1.6	1.9	0.4	1.9	-	-	0.02-0.5
SO ₃ (%)	6.7	7.0	3.0	4.7	5.5	2.2	1.9	1.8-4.6	0.1	0.2-22.1
TiO ₂ (%)	1.4	1.4	0.9	1.3	1.4	0.6	1.0	-	-	0.23-1.58
Cl (%)	13	0.6	23	3	6.5	27	3.8	-	-	-

Table 4 Major components of the fly ashes before and after ED treatment. ¹ Belmonte et al. 2016, ² Kosmatha et al. 2011, ³ Haiying et al. 2011, ⁴Lin, 2006, ⁵Jayaranjan et al. 2014