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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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Abstract

In Greenland, fly ash could contribute as a local resource in construction as a substitute for cement in concrete or clay in bricks, if the toxicity of the ash is reduced. In this study, fly ash from three different Greenlandic waste incinerators were collected and subjected to electrodialytic treatment 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 removal was mostly dependent on the initial concentration in the fly ash. Heavy metal leaching was 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 purposes. Hg leaching was also reduced to below Danish guideline levels, although only investigated for one fly ash. Hexavalent Cr was not the dominant speciation of Cr in the fly ashes. Ettringite formed during electrodialytic treatment in the fly ash suspensions at pH above 12. The total concentration of eligible components for reuse such as CaO, SiO₂ and Al₂O₃, increased during the electrodialytic treatment.

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

22 **1. Introduction**

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

One method that has shown potential for treating incineration residues is electrodialytic remediation (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 has been made in laboratory scale, where suspending the residues in distilled water have been found 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, 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. The principle in ED treatment is shown in Fig. 1. The ED system consists of an ED stack with multiple concentrate compartments (concentrate) and compartments containing the fly ash suspension. Ion exchange membranes separate the compartments and the setup is designed to remove anions and cations from the fly ash suspension to the concentrate by the applied current. 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-splitting at the anion exchange membrane (Ottosen et al. 2000) or assisting agents. This acidification implies long treatment times. Removals up to 90% Cd, 85% Cu, 60 % Zn, 45 % Cr 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 alters the treated material's matrix significantly. This is seen by dissolution of up to 78 % fly ash 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 12 % when using assisting agents (Pedersen et al., 2005). The later ED experiments in bench - and pilot scale focused on the reduction of the metal and salt leaching to reduce environmental hazardousness while leaving the non-mobile fractions in the ED treated material. Thus, larger

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

79 Solid waste has typically been dumped at uncontrolled landfills in the Arctic (Poland et al., 2003).
80 Use of uncontrolled landfills was also the practice in Greenland before first waste incinerator started
81 operating the 1990ies, primarily to limit the amount of waste for disposal and hygienize the waste.
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
84 European waste incinerators and have a capacity between 2,000 – 10,000 t annually (Eisted and
85 Christensen, 2011) and represent a waste management option for smaller isolated communities. The
86 grate furnace incinerators in Nuuk, Sisimiut and Qaqortoq have flue gas treatment only in the form
87 of electrostatic precipitators (ESP) and the grate furnace incinerators in Maniitsoq, Ilulissat and
88 Aasiaat have cyclones followed by electrostatic precipitators. This equipment is capable of
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
91 any of the Greenlandic incinerators. All incinerators have heat boilers for energy recovery. The
92 resulting residues from the incinerators are bottom ash, boiler ash and fly ash. The boiler and

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

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

115

116 2. Materials and methods

117 2.1. Experimental fly ashes

118 ESP fly ashes (without acid gas cleaning products) were collected from big bags at the temporary
119 storages at random days from the incinerators in Nuuk, Sisimiut and Qaqortoq, which have annual
120 incineration capacities of 10,000; 4,000 and 3,000 tons respectively (Eisted and Christensen, 2011).
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
123 work. The fly ashes were sieved through a 1 mm sieve before the electrodialytic experiments not to
124 block the ED stack. For the Sisimiut and Qaqortoq fly ashes the amount passing the 1 mm sieve was
125 100 % and for the Nuuk fly ash, 92 % passed the 1 mm sieve.

126 2.2. Analytical procedures

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

139 electrode. The amount of water-soluble fly ash was estimated as mass reduction when mixing 1 g
140 fly ash with 20 mL of distilled water which was agitated for 24 hours. pH-dependent batch
141 extraction experiments were made with HNO_3 and as a reference distilled water was used. The
142 extractions were made by mixing 10.0 g fly ash with 50 mL HNO_3 in concentrations of 0.01 M –
143 0.7 M in 100 mL acid rinsed plastic bottles. For the Qaqortoq sample, NaOH in concentrations of
144 0.02 – 0.05 M was also added to increase the pH. All the extractions were made in triplicates and
145 shaken for 72 hours on a horizontal shaker operating at 250 rpm to keep the fly ash in suspension.
146 After that the samples settled for 15 minutes before pH was measured. The extractants were filtrated
147 through a 45 μm filter and acidified if $\text{pH} > 2$ by addition of concentrated HNO_3 before
148 measurements of metals by ICP-OES. Extractions in triplicate on solid fly ash samples for Cr (VI)
149 total analysis were made according to the US EPA 3060A method. Cr (VI) in the solutions was
150 measured by colourimetrically UV-VIS spectrophotometry on a Spectroquant Nova 60.

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 $\text{K}\alpha$ 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**

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

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

169

170 The ED stack consisted of 5 cell pairs (530 cm² active surface membrane area per cell) and
171 Neosepta cation and anion exchange membranes were used. The spacers were 0.6 mm and 5 mm
172 thick for the concentrate and fly ash suspension compartments respectively. The spacers used for
173 the fly ash suspension were thicker than the concentrate spacers to avoid clogging due to the high
174 solids content in the fly ash suspension. In Fig. 1 it is seen how the ion exchange membranes and
175 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
178 ED stack, resulting in current densities of 9.4 or 4.7 mA/cm², by 530 cm² active surface membrane
179 area. The electrode compartments shared electrode solution, which was circulating. The pumps
180 circulating the concentrate, fly ash suspension and electrode solution were for experiments N-1 and
181 N-2 centrifugal pumps. For the remaining experiments, the pump for the fly ash suspension was a
182 peristaltic pump PSF3 from Ragazzini. The concentrate and electrolyte solutions were 5 L each of
183 distilled water adjusted to pH 1 with HNO₃ to avoid precipitation of metals in the pumps, spacers
184 and electrodes. The fly ash suspensions were made by mixing 1 - 2.5 kg fly ash with 10 L distilled

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

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

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

$$\text{Removal (\%)} = ((\text{CL}_{\text{concentrate}} \times \text{V}_{\text{Concentrate}} + \text{CL}_{\text{electrode solution}} \times \text{V}_{\text{Concentrate solution}}) / \text{C}_{\text{fly ash}} \times \text{M}_{\text{fly ash}}) \times 100$$

200 %

201 Where CL (mg/L) is the measured concentration in the concentrate and electrode solution, V (L) the
202 volume, C (mg/kg) the concentration in the fly ash and M (kg) the initial mass of fly ash in the
203 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
207 incineration plants have the same flue gas treatment systems, see Table 2. The Nuuk and Sisimiut
208 fly ashes have the characteristic alkaline pH of MSWI fly ash, but the Qaqortoq fly ash had a
209 neutral pH. This lower pH of the latter fly ash is not unusual (Hjelmar et al., 2011) and could be due
210 to the technical performance of the fly ash collection and cooling of the flue gas in the incineration
211 plant. The composition of the fuel and the combustion process are important factors for
212 determining the heavy metal content in the fly ash, especially Cl influences the amount of Pb in the
213 fly ash (Pedersen et al., 2009). Regardless, the total concentrations of Cd, Cr, Cu, Pb and Zn were in
214 the same orders of magnitude for the three fly ashes. The fraction of Cr determined as Cr (VI) was
215 low compared to the total Cr concentration in all three ashes and Cr (VI) was not the dominant
216 speciation of Cr in the fly ashes before the ED. These results show that the ESP ash from the small-
217 scale Greenlandic incinerators was within the typical ranges of fly ash from larger incineration
218 plants (Lam et al., 2010). Fig. 2 shows the pH-dependent desorption of the heavy metals. It can be
219 seen that the pH-dependent desorption followed a similar pattern for the metals in the three different
220 ashes. Cd, Cu and Zn were desorbed at acidic pH mainly, whereas Cr and Pb to some extent also
221 were desorbed at alkaline pH. Zn desorption from the Nuuk ash started below pH 4 and was
222 desorbed to a lower degree than for the Sisimiut and Qaqortoq ashes. Generally, the maximum
223 desorption of the metals (~100 %) was seen at pH 2, although the desorption degree from the Nuuk
224 ash was between 70-80 % for the metals. The different fractions that fly ash typically consist of are
225 soluble in different pH ranges; oxides between $2 < \text{pH} < 6$, carbonates $6 < \text{pH} < 9$ and hydroxides
226 $10 < \text{pH} < 13$ (Flyvbjerg and Hjelmar, 1997). The release of metals in these pH ranges could
227 indicate preferable speciation of the metals towards these phases. Compared to the experimental
228 results in Fig. 2, the highest desorption was seen in the pH range from 0 to 7, indicating dominating
229 oxide speciation, and for Cd and Zn, also carbonates, as desorption was seen around pH 7. A

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).

3.2. Electrodialytic experiments

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

The voltage drop was not directly linked to the electrical conductivity in the fly ash suspension, so the high resistance resulting in the maximum voltage of the power supply in experiments N-1 and 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 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 system were seen with L/S 5 and 4. The electrical conductivity in the fly ash suspension decreased with time due to the continuous removal of ions in the stack system. pH in the fly ash suspensions decreased only slightly throughout the experiments, which was expected due to the buffering 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 much higher than that of harbour sediments.

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
254 for Cd, Cu and Zn was seen in the Sisimiut and Qaqortoq fly ash suspension, which had lower pH
255 than the Nuuk fly ash suspension. The highest removals were 15 % Cd and 49 % Cu in the Q-1
256 experiment and 7.4 % and 10.1 % Pb in N-1 and N-2. All other removal rates were below 6 %.

257 Previous work has shown removals of up to 55 % Cd, 4.5 % Cr, 90 % Cu, 66 % Pb and 55 % Zn
258 from fly ash in water suspensions, which reached final pH 3.9 – 8.2 in electrodialytic experiments
259 (Ottosen et al., 2006, Kirkelund et al., 2015). This pH was only achieved in the Q-1 experiment,
260 where the removals were 15 % Cd, 49 % Cu, 2 % Cr, 3 % Pb and 5 %. The removals do not take
261 into account any amount of metal that has electrodeposited on the cathode, which readily occurs for
262 Cu and Pb (Ottosen et al., 2003, Ottosen et al., 2006). The efficiency of electrodeposition of metal
263 cations is dependent on several factors such as electrode material, current density, electrode surface
264 area and the electrolyte solution matrix (Chen, 2004) and could underestimate the removals in this
265 study. Lima et al. (2010) showed that ash type, duration of the electrodialytic experiment and
266 dissolution of fly ash during electrodialytic remediation to be the most important factors for metal
267 removal. The duration of the experiments was short, however, the applied current higher than in
268 previous experiments.

269 The amount of metal found in the filtered suspension liquid indicates released metal compounds
270 that are uncharged and which cannot electromigrate and stay in the suspension. The highest amount
271 found in the suspension was for Cr in all experiments except N-1 and N-2 and Pb in the N-1 and N-
272 2 experiments. Pb forms stable complexes with Cl and Pedersen (2002) predicted that in a fly ash
273 solution with a high concentration of Cl, 35 % of Pb would be present as uncharged PbCl_2 . Also at
274 the high pH in the Nuuk experiments, $\text{Pb}(\text{OH})_2$ could be a dominant species. Thus, a lower pH
275 favours a larger fraction of charged Pb species, which is removed in the electric field as seen in all
276 Sisimiut and the Q-1 experiments.

277 The amount of Cr in the suspension liquid was significant, regardless of pH in the solution and
278 operational conditions. Thus, the speciation and behaviour of Cr in the ED stack differ significantly
279 to the other investigated metals. Hexavalent Cr would be on anionic form and electromigrate
280 towards the anode and trivalent Cr on cationic form and electromigrate towards the cathode.
281 However, with the combined electrode solution and the concentrate, it is not possible in this
282 experimental setup to determine the charge of the removed Cr. There also seems to be a dependence
283 on the type of fly ash, as a similar amount of Cr in the suspension liquid has not been seen for
284 Danish fly ash nor APC residue (Kirkelund et al., 2015). $\text{Cr}(\text{OH})_3$ could be the dominant species in
285 the solution in the pH range in the experiments (pH 7-12) (Ščančar and Milačič, 2014). Although
286 the Cr was found in the suspension liquid, it is released from the fly ash particles, and this is
287 consistent with results from the pH-dependent desorption (Fig. 2), which showed a high release of
288 Cr at alkaline pH.

289 **3.4. Heavy metal content and leaching**

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

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

316 Cr leaching decreased due to the ED treatment for all Sisimiut experiments and experiment N-2,
317 contrarily to what has been seen for Danish APC residues and fly ash where the Cr leaching always
318 increased (Jensen et al., 2015; Kirkelund et al., 2010; Kirkelund et al., 2015). The total Cr
319 concentrations in the solid fly ash increased for all fly ashes except S-1 and S-4 and the hexavalent
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
322 treatment. For most fly ash samples, the Cr (VI) measurement was below the detection limit of 2
323 mg/kg. Based on the L/S ratio for the analysis this corresponds to a leached Cr value of 1 mg/l, and

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

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

342 The treatment methods are divided into washing with different liquids, thermal methods,
343 electrokinetic and as combinations, as several treatment methods are often used together. Several
344 studies apply treatment methods before a valorisation test in construction material; however, only a
345 few of these studies give information about leaching of the treated fly ash itself. They rather present
346 the leaching results from the construction material into which the treated fly ash was incorporated,
347 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, which implies the difficult nature of the fly ashes, as shown in Fig. 5. Also, some leaching tests were made with the TCPL test using acetic acid instead of distilled water according to the DS/EN 12457-3 leaching test commonly applied in Europe. The leaching results with the TCPL tests are 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. Very few of the methods from the literature make the leaching stand out as particularly low or below the limits they are compared with for all metals studied. As the heavy metal leaching is highly pH dependent and each treatment method result in different pH of the MSWI fly ash, the leaching is highly dependent on final pH. Although, a combination of treatments seems to be the most viable option, commonly for all methods is a significant decrease of the initial leaching of the fly ashes (Chen et al., 2017; Liao et al., 2014; Liu et al., 2009; Lundtorp et al., 2002) which is beneficial for valorization. As an oxyanion Cr behaves differently from other metals and even increased leaching of Cr has been observed when fly ash is treated by thermal treatment or water washing (Liu et al., 2009; Lundtorp et al., 2002). This increased leaching can be due to oxidation of trivalent Cr to hexavalent Cr at high temperature (Wang et al. 2001) and an increase in the soluble fraction of Cr as calcium chromate when in reaction with CaO and CaCl₂ during thermal treatment (Kirk et al., 2002). When ash is in contact with water, hydroxide ions are formed by hydrolysis of metal oxides, and this increases the pH, which favours Cr leaching (Nordmark and Lagerkvist, 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 Lagerkvist, 2018). It is, therefore, necessary to control pH and redox conditions in the fly ash to reduce the Cr leaching.

3.5 Matrix changes

372 The XRD diffractograms are seen in Fig. 6 from where the major crystalline phases in the fly ashes
 373 before and after ED treatment are specified. The major components in the fly ashes before and after
 374 ED treatment are shown in Table 4 and compared to major components in cement, clay and coal fly
 375 ash (CFA). The untreated fly ashes consisted mainly of Ca and Cl, and Na, K and Al to a lower
 376 degree (Table 4). The main mineral phases of the fly ashes were also linked to these elements and
 377 classified as Ca-containing minerals such as anhydrite (CaOH) and carbonate (CaCO_3) and Cl-
 378 containing minerals such as sylvite (KCl) and halite (NaCl). Formation of ettringite
 379 ($\text{Ca}_6\text{Al}_2(\text{SO}_4)_3 \cdot 26\text{H}_2\text{O}$), which is an indication of ageing of the ash (Steenari et al., 1999) were
 380 seen in both the Nuuk ED experiments and the S-2 experiment, but not in the other fly ash samples
 381 after ED treatment. The formation of gypsum ($\text{CaSO}_4 \cdot \text{H}_2\text{O}$) was significant for the Sisimiut and
 382 Qaqortoq fly ash during ED treatment as opposed to the Nuuk fly ash. Formation of ettringite rather
 383 than gypsum is dependent on pH and has found to be favoured at pH between 11 and 12.5
 384 (Chrysochoou and Dermatas, 2006), which also coincides with the pH of the Nuuk fly ash
 385 suspensions as seen in Fig. 3. The Zn-containing mineral simonkolleite ($\text{Zn}_5(\text{OH})_8\text{Cl}_2 \cdot \text{H}_2\text{O}$) was
 386 identified for the Qaqortoq fly ash after ED treatment, which corresponds well to the high Zn
 387 concentration after the EDR treatment. During ED treatment soluble minerals such as the Cl-
 388 minerals dissolved and could be removed in the electric field. Na, K and Cl were removed from the
 389 fly ashes, but to a greater extent for the Nuuk ash than Sisimiut and Qaqortoq, which could be due
 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
 392 treated ash. Thus, increasing the treatment time increased the Cl removal, which was also seen in
 393 another study by (Kirkelund et al., 2015) in laboratory scale EDR experiments.

394 CaO , SiO_2 and Al_2O_3 are major components that are desirable for the valorisation of fly ash as
 395 substitution of primary resources in concrete, glass-ceramics, ceramic products and glass materials

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

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

413 **4. Conclusion**

414 The composition of Greenlandic fly ash varied among the three investigated incinerators, even
415 though the flue gas treatment systems were similar. The heavy metal removal was more dependent
416 on the fly ash and initial heavy metal content rather than the experimental conditions such as current
417 density, duration and L/S ratio. Cr was not removed from the liquid phase of the fly ash suspension
418 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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435 **References**

- 436 Aubert, J.E., Husson, B., Sarramone, N., 2006. Utilization of municipal solid waste incineration
437 (MSWI) fly ash in blended cement Part 1: Processing and characterization of MSWI fly ash. J.
438 Haz. Mat. 136, 624-631
- 439 Belmonte, L.J., Ottosen, L.M., Kirkelund, G.M., Jensen, P.E., Vestbø, A.P., 2016. Screening of
440 heavy metal containing waste types for use as raw material in Arctic clay-based bricks. Environ
441 Sci Pollut Res, doi:10.1007/s11356-016-8040-z

442 Boesch, M.E., Vadenbo, C., Saner, D., Huter, C., Hellweg, S., 2014. An LCA model for waste
 443 incineration enhanced with new technologies for metal recovery and application to the case of
 444 Switzerland. *Waste Manag.* 34, 378–389. doi:10.1016/j.wasman.2013.10.019
 445 Buonanno, G., Morawska, L., 2015. Ultrafine particle emission of waste incinerators and
 446 comparison to the exposure of urban citizens. *Waste Manag.* 37, 75–81.
 447 doi:10.1016/j.wasman.2014.03.008
 448 Chen, G., 2004. Electrochemical technologies in wastewater treatment. *Separation and Purification*
 449 *Technology* 38, 11-41.
 450 Chen, W., Kirkelund, G.M., Jensen, P.E., Ottosen, L.M., 2017. Combined water washing and
 451 electrodialytic treatment of MSWI fly ash - Effect on As , Cr , Pb and Zn leaching after
 452 thermal treatment. *Waste Manag.* doi:10.1016/j.wasman.2017.07.011
 453 Chen, W., Kirkelund, G.M., Jensen, P.E., Ottosen, L.M., 2018. Electrodialytic extraction of Cr from
 454 water-washed MSWI fly ash by changing pH and redox conditions. *Waste Management* 71,
 455 215-223.
 456 Chrysochoou, M., Dermatas, D., 2006. Evaluation of ettringite and hydrocalumite formation for
 457 heavy metal immobilization: Literature review and experimental study. *J. Hazard. Mater.* 136,
 458 20–33. doi:10.1016/j.jhazmat.2005.11.008
 459 Chu, T.-C., Wang, K.-S., Lin, K.-L., Chien, C.-C., Chen, J.-H., 2013. Synthesis of waste-derived
 460 glass-ceramics from MSWI fly ash and EAF dust: Kinetics of nucleation and crystallization.
 461 *Environ. Prog. Sustain. Energy* 32, 480–488. doi:10.1002/ep.11647
 462 Eisted, R., Christensen, T.H., 2011. Waste management in Greenland: current situation and
 463 challenges. *Waste Manag. Res.* 29, 1064–1070. doi:10.1177/0734242X10395421

464 EPA 2004. Chapter Seven of the SW-846 Compendium: Introductory and Regulatory Definitions
 465 Pertaining to Hazardous Waste Characteristics, Revision 4, November 2004.

466 Ferreira, C., Ribeiro, A., Ottosen, L., 2003. Possible applications for municipal solid waste fly ash.
 467 J. Hazard. Mater. 96, 201–216. doi:10.1016/S0304-3894(02)00201-7

468 Flyvbjerg, J., Hjelm, O., 1997. Restprodukter fra røggasrensning ved affaldsforbrænding 3.
 469 Arbejdsrapport fra Miljøstyrelsen. VKI og Miljø-og energiministeriet nr. 92.

470 Gilbreath, S., Krass, P.H., 2006. Adverse Birth Outcomes Associated with Open Dumpsites in
 471 Alaska Native Villages. Am. J. Epidemiol. 164, 518–528. doi:10.1093/aje/kwj241

472 Haiying, Z., Youcai, Z., Jingyu, Q., 2011. Utilization of municipal solid waste incineration (MSWI)
 473 fly ash in ceramic brick: Product characterization and environmental toxicity. Waste Manag.
 474 31, 331–341. doi:10.1016/j.wasman.2010.10.017

475 Hjelm, O., Johnson, A., Ash, B., 2011. Incineration: Solid Residues. In: Solid Waste Technology
 476 & Management. Blackwell, Chichester, UK.

477 Jayaranjan, M.L.D., van Hullebusch, E.D., Annachatre, A.P., 2014. Reuse options for coal fired
 478 power plant bottom ash and fly ash. Rev. Environ. Sci. Bio/Technology 467–486.
 479 doi:10.1007/s11157-014-9336-4

480 Jensen, P.E., Ferreira, C.M.D., Hansen, H.K., Rye, J.-U., Ottosen, L.M., Villumsen, A., 2010.
 481 Electroremediation of air pollution control residues in a continuous reactor. J. Appl.
 482 Electrochem. 40, 1173–1181. doi:10.1007/s10800-010-0090-1

483 Jensen, P.E., Kirkelund, G.M., Pedersen, K.B., Dias-Ferreira, C., Ottosen, L.M., 2015.
 484 Electrodialytic upgrading of three different municipal solid waste incineration residue types

485 with focus on Cr, Pb, Zn, Mn, Mo, Sb, Se, V, Cl and SO₄. *Electrochim. Acta* 181.
 486 doi:10.1016/j.electacta.2015.06.012

487 Jones, A.M., Harrison, R.M., 2016. Emission of ultrafine particles from the incineration of
 488 municipal solid waste: A review. *Atmos. Environ.* 140, 519–528.
 489 doi:10.1016/j.atmosenv.2016.06.005

490

491 Kirk, D.W., Chan, C.C.Y., Marsh, H., 2002. Chromium behavior during thermal treatment
 492 of MSWfly ash, *J. Hazard. Mater.* 7 39–49.

493 Kirkelund, G.M., Jensen, P.E., Villumsen, A., Ottosen, L.M., 2010. Test of electrodialytic
 494 upgrading of MSWI APC residue in pilot scale: Focus on reduced metal and salt leaching. *J.*
 495 *Appl. Electrochem.* 40. doi:10.1007/s10800-009-0059-0

496 Kirkelund, G.M., Magro, C., Guedes, P., Jensen, P.E., Ribeiro, A.B., Ottosen, L.M., 2015.
 497 Electrodialytic removal of heavy metals and chloride from municipal solid waste incineration
 498 fly ash and air pollution control residue in suspension - Test of a new two compartment
 499 experimental cell. *Electrochim. Acta* 181. doi:10.1016/j.electacta.2015.03.192

500 Kosmatka, S.H., Wilson, M.L., 2011. Design and control of concrete mixtures, EB001 15th edition.
 501 Portland Cement Association, Skokie, Illinois, USA.

502 Lam, C.H.K., Ip, A.W.M., Barford, J.P., McKay, G., 2010. Use of incineration MSW ash: a review.
 503 *Sustainability* 2, 1943-1968.

504 Liao, W.-P., Yang, R., Zhou, Z.-X., Huang, J.-Y., 2014. Electrokinetic stabilization of heavy metals
 505 in MSWI fly ash after water washing. *Environ. Prog. Sustain. Energy* 33, n/a-n/a.

506 doi:10.1002/ep.11920

507 Lima, A. T., Ottosen, L.M., Pedersen, a. J., Ribeiro, a. B., 2008. Characterization of fly ash from
508 bio and municipal waste. *Biomass and Bioenergy* 32, 277–282.

509 doi:10.1016/j.biombioe.2007.09.005

510 Lima, A.T., Ottosen, L.M., Ribeiro, A.B., 2012. Assessing fly ash treatment: Remediation and
511 stabilization of heavy metals. *J Environ Management* 95, S110-S115.

512 Lin, K.L., 2006. Feasibility study of using brick made from municipal solid waste incinerator fly
513 ash slag. *J Haz Mater B* 137, 1810-1816.

514 Liu, Y., Zheng, L., Li, X., Xie, S., 2009. SEM/EDS and XRD characterization of raw and washed
515 MSWI fly ash sintered at different temperatures. *J. Hazard. Mater.* 162, 161–173.

516 doi:10.1016/j.jhazmat.2008.05.029

517 Lundtorp, K., Jensen, D.L., Sørensen, M. a, Christensen, T.H., Mogensen, E.P.B., 2002. Treatment
518 of waste incinerator air-pollution-control residues with FeSO₄: concept and product
519 characterisation. *Waste Manag. Res.* 20, 69–79. doi:10.1177/0734242X0202000108

520 Margallo, M., Taddei, M.B.M., Hernández-Pellón, A., Aldaco, R., Irabien, Á., 2015. Environmental
521 sustainability assessment of the management of municipal solid waste incineration residues: A
522 review of the current situation. *Clean Technol. Environ. Policy* 17, 1333–1353.

523 doi:10.1007/s10098-015-0961-6

524 Miljøstyrelsen, 2011. Bekendtgørelse om deponeringsanlæg. BEK nr 719 af 24/06/2011, 2011 (in
525 Danish).

526 Miljøstyrelsen, 2015. Bekendtgørelse om genanvendelse af restprodukter og jord til bygge-og

527 anlægsarbejder (Restproduktbekendtgørelsen) BEK nr 1414 af 30/11/2015 (in Danish).

528 Nordmark., D., Lagerkvist, A., 2018. Controlling the mobility of chromium and molybdenum in
 529 MSWI fly ash i a washing process. *Waste Management* 76, 727-733.

530 Ottosen, L.M., Kristensen, I.V., Pedersen, A.J., Hansen, H.K., Villumsen., A., Ribeiro, A.B., 2003.
 531 Electrodialytic removal of heavy metals from different solid waste products. *Separation*
 532 *Science and Technology* 38 (6), 1269-1289.

533 Ottosen, L.M., Hansen. H.K., Hansen, C., 2000. Water splitting at ion-exchange membranes and
 534 potential differences in solid during electrodialytic remediation. *J. Appl. Electrochem.* 30,
 535 1199-1207.

536 Ottosen, L.M., Lima, A.T., Pedersen, A.J., Ribeiro, A., B., 2006. Electrodialytic extraction of Cu,
 537 Pb and Cl from municipal solid waste incineration fly ash suspended in water. *J. Chem.*
 538 *Technol. Biotechnol.* 81, 553-559.

539 Pedersen, A.J., 2002. Evaluation of assisting agents for electrodialytic removal of Cd, Pb, Zn, Cu
 540 and Cr from MSWI fly ash. *J. Hazard. Mater.* 95, 185–198. doi:10.1016/S0304-
 541 3894(02)00138-3

542 Pedersen, A.J., Frandsen, F.J., Riber, C., Astrup, T., Thomsen, S.N., Lundtorp, K., Mortensen, L.F.,
 543 2009. A full-scale study on the partitioning of trace elements in municipal solid waste
 544 incinerations-Effects of firing different waste types. *Energy and Fuels* 23, 3475–3489.
 545 doi:10.1021/ef801030p

546 Pedersen, A.J., Gardner, K.H., 2003. Characterization of municipal solid waste incineration fly ash
 547 before and after electrodialytic treatment. *J. Phys. IV JP* 107, 1029–1032.

548 Pedersen, K.B., Ottosen, L.M., Jensen, P.E., Lejon, T., 2015. Comparison of 2-compartment, 3-
549 compartment and stack designs for electrodialytic removal of heavy metals from harbour
550 sediments. *Electrochim. Acta* 181, 48–57. doi:10.1016/j.electacta.2014.12.003

551 Poland, J.S., Riddle, M.J., Zeeb, B.A., 2003. Contaminants in the Arctic and the Antarctic: a
552 comparison of sources, impacts, and remediation options. *Polar Rec. (Gr. Brit)*. 39, 369–383.
553 doi:10.1017/S0032247403002985

554 Quina, M.J., Bordado, J.C., Quinta-Ferreira, R.M., 2008. Treatment and use of air pollution control
555 residues from MSW incineration: an overview. *Waste Manag.* 28, 2097–121.
556 doi:10.1016/j.wasman.2007.08.030

557 Rémond, S., Pimienta, P., Bentz, D., 2002. Effects of the incorporation of Municipal Solid Waste
558 Incineration fly ash in cement pastes and mortars I. Experimental study. *Cem. Concr. Res.* 32,
559 303–311. doi:10.1016/S0008-8846(01)00674-3

560 Ščančar, J., Milačič, R., 2014. A critical overview of Cr speciation analysis based on high
561 performance liquid chromatography and spectrometric techniques. *J. Anal. At. Spectrom.* 29,
562 427. doi:10.1039/c3ja50198a

563 Steenari, B.M., Schelander, S., Lindqvist, O., 1999. Chemical and leaching characteristics of ash
564 from combustion of coal, peat and wood in a 12MW CFB – a comparative study. *Fuel* 78,
565 249–258. doi:10.1016/S0016-2361(98)00137-9

566 Todorovic, J., Ecke H., 2006. Treatment of MSWI Residues for Utilization as Secondary
567 Construction Minerals: A Review of Methods. *Minerals and Energy-Raw Materials Report* 20, 45-
568 59.

569 Viader, R.P., Jensen, P.E., Ottosen, L.M., 2017. Electrodialytic remediation of municipal solid
570 waste incineration residues using different membranes. *Chemosphere* 169, 62-68.

571 Wang, K.S., Sun, C.J., Liu, C.Y., 2001. Effects of the type of sintering atmosphere on the
572 chromium leachability of thermal-treated municipal solidwaste incinerator fly
573 ash. *Waste Management*, 21 85–91.

574 Weichenthal, S., Van Rijswijk, D., Kulka, R., You, H., Van Ryswyk, K., Willey, J., Dugandzic, R.,
575 Sutcliffe, R., Moulton, J., Baike, M., White, L., Charland, J.P., Jessiman, B., 2015. The impact
576 of a landfill fire on ambient air quality in the north: A case study in Iqaluit, Canada. *Environ.*
577 *Res.* 142, 46–50. doi:10.1016/j.envres.2015.06.018

578 Weiser, E.L., Powell, A.N., 2011. Reduction of Garbage in the Diet of Nonbreeding Glaucous Gulls
579 Corresponding to a Change in Waste Management. *ARCTIC* 64. doi:10.14430/arctic4101

580 Zacco, A., Borgese, L., Gianoncelli, A., Struis, R.P.W.J., Depero, L.E., Bontempi, E., 2014. Review
581 of fly ash inertisation treatments and recycling. *Environ. Chem. Lett.* 12, 153–175.
582 doi:10.1007/s10311-014-0454-6

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

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

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. 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 (hours)	Applied current (A)	Current density (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

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

Parameter	Nuuk	Sisimiut	Qaqortoq	ESP fly ash ¹
pH	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 3 Leaching and total concentrations in the fly ash samples before and after electrodialytic treatment. ¹Miljøstyrelsen 2015, ²Jensen et al. 2015.

		Nuuk	N-1	N-2	Sisimiut	S-1	S-2	S-3	S-4	Qaqortoq	Q-1	C3 ¹	CFA ²
Total content	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
	Cr (VI) (mg/kg)	3.8	<2	<2	<2	<2	2.7	4.9	<2	2.2	2.5	>20	-
	Cu (mg/kg)	718	1,310	1,140	1,540	1,530	2,080	2,160	1,770	713	1,680	>500	32
	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
Leaching	pH	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
	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
	Cu(µg/l)	300	61	41	74	65	76	80	80	60	<20	2000	75
	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 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

	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 equivalent)	0.5	0.04-1.23
K ₂ O (%)	6.5	0.2	8.9	2.0	3.7	12.0	2.4		2.7-3.1	0.2-4.89
Al ₂ O 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 ₂ O 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 ₂ O 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	-	-	-