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CHARACTERIZATION OF PARTICULATE RESIDUES FROM GREENLANDIC MSWI FOR USE AS SECONDARY RESOURCES

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Abstract
In Greenland, waste incineration is used in the larger towns to treat the municipal solid waste. The incineration reduces the amount of waste, but produces particulate incineration residues such as fly and bottom ash that are disposed of. Most construction materials are imported to Arctic areas. The focus in this study is a characterisation of Greenlandic incineration residues to assess the potential as local secondary resources. In this study, fly ash samples from all the incinerators and bottom ash from two incinerators were collected and investigated for several physical-chemical properties. The fly ash samples consisted of very fine-grained particles, with different grading for each incinerator. High water solubility due to high salt concentrations was seen for all fly ash samples along with high concentrations of leachable heavy metals, thus pre-treatment is recommended before use as secondary material. The bottom ashes consisted of coarser particles and exhibited lower heavy metal leaching than the fly ash. All residue samples were different and evaluation of reuse should be made individually, however the fly ash shows potential as cement replacement and bottom ash as sand replacement for construction purposes.

1. Introduction

Materials and consumer products are usually shipped long distances during the summer to remote Arctic settlements from the southern regions of the world. After the end use of these materials and products end up as waste, which will not be shipped back but has to be managed locally. Solid waste has typically been dumped at uncontrolled landfills in the Arctic and the waste is accumulating. These uncontrolled landfills rise problems such as uncontrolled burning, contamination of surface and groundwater and emission of greenhouse gases. This lack of waste

management is mainly due to small populations which traditionally have created limited amounts of waste, access to large areas away from the inhabitants, low tax base population and limited environmental awareness.

Municipal solid waste incineration (MSWI) was implemented in Greenland as part of the country's first waste strategy. The first waste incinerator started operating in the capital Nuuk in 1989 and waste incinerators followed in five other towns (Aasiaat, Ilulissat, Maniitsoq, Qaqortoq and Sisimiut) in the 2000s, primarily to limit the amount of waste for disposal and hygenize the waste. The Greenlandic incineration plants are smaller and simpler than traditional European waste incinerators and have a capacity between 2,000 – 10,000 t annually [1] which is a typical daily capacity in larger European incinerators.

The incinerators in Nuuk, Sisimiut and Qaqortoq have flue gas treatment in the form of electrostatic precipitators (ESP) and the incinerators in Maniitsoq, Ilulissat and Aasiaat have cyclones followed by an electrostatic precipitator. This equipment is only capable of removing particulate material and so gaseous contaminants such as volatilized metals and acids are emitted to the atmosphere. Flue gas treatment for organic pollutants such as PAHs and dioxins also do not exist. All incinerators have heat boilers for energy recovery and the incineration temperature is maximum 1000 °C. The resulting residues from the incineration are bottom ash, boiler ash and fly ash after the flue gas treatment. The annual production of fly ash and bottom ash from the Greenlandic incinerators are 6,000 t MSWI bottom ash and 200 t MSWI fly ash [1]. The boiler and bottom ashes are collected together and disposed of at the uncontrolled disposal sites. The fly ashes are stored at the disposal sites before being shipped for disposal in Norway, via Denmark. This shipment and safe disposal is a considerate expense for the Greenlandic municipalities. The environmental impact from disposal of fly ashes in the current Greenlandic disposal sites is considered too high for local disposal to be an alternative.

Simultaneously, all construction materials are shipped to Greenland from Denmark, as the only used local construction materials in Greenland are primary resources as sand and rock. The value for import of materials and goods for construction varied from 55 to 62 million euros for the years 2010-2015, which is around 10 % of the value of the total import to Greenland [2]. Incineration residues are considered potential valuable secondary resources and could be used in construction or geotechnical applications [3 ,4], and the Greenlandic incineration residues could have potential as secondary resources.

In this study, fly ash samples from all the six Greenlandic incinerators and bottom ash from two of the incinerators were collected and investigated for several physical-chemical properties to evaluate the similarities between the ashes from the different incineration plant and potential reuse options of the incineration residues.

2. Materials and methods

2.1 Experimental fly ashes

The fly ash was collected from the big bags, where the ash is collected after the electrostatic precipitators at a random day in 2010 from the incinerators in Nuuk, Sisimiut, Ilulissat, Aasiaat,

Maniitsoq and Qaqortoq. For Aasiaat a separate sample of cyclone ash (CA) was also collected. There is no information about the specific type of waste incinerated on the days of sampling. The Nuuk incinerator has a capacity of 1600 kg/h and the other incinerators have a capacity of 400-800 kg/h. There are three main types of incinerators; Nuuk (B&S furnace); Aasiaat, Ilulissat and Maniitsoq (Weiss furnace); Qaqortoq and Sisimiut (REKA furnace). Bottom ash (BA) from the incinerator in Sisimiut and Ilulissat was collected directly from the containers where the bottom ash is collected, prior to disposal. The ashes were stored in sealed boxes at room temperature prior to the experimental work.

2.2 Analytical procedures

Heavy metal concentrations (As, Ba, Cd, Cr, Cu, Mn, Ni, Pb, Zn) in the ashes were measured by ICP-OES after pre-treatment by Danish Standard DS259 where 1 g of ash and 20 ml 7.3 M HNO_3 were heated at 200 kPa (120°C) for 30 min. The liquid was thereafter separated by vacuum filtration through a 45 µm filter and diluted to 100 ml. The units used in this paper are mg/kg for concentrations in dry matter and the result is the average of at least three samples. The heavy metals were measured by induced coupled plasma – optical emission spectrometry (ICP-OES).

Water content was measured by weight loss at 105°C for 24 h. Loss on ignition was measured at 550°C for one hour. The pH was measured in 1 M KCl at a liquid-to-solid ratio (L/S) of 5 and after 1 hour of agitation, pH was measured by a Radiometer Analytical pH electrode. The amount of water soluble fly ash was estimated as mass reduction when mixing 1 g fly ash with 20 ml distilled water which was agitated for 24 hours.

Leaching experiments were made according to CEN prEN 12457-3 part 1. The liquid to solid (L/S) ratio was L/S 2, mixing 40 g ash and 80 ml distilled water. The suspension was shaken for 6 hours on an end-over shaker before vacuum filtration through a 45 µm filter and the filtrate was divided into two subsamples. The sample was then acidified by addition of concentrated HNO_3 before analysis of heavy metals by ICP-OES.

Major oxide composition of the fly ashes was estimated from semi-quantitative analysis by X-ray fluorescence (XRF) on powder samples by an external laboratory. Scanning electron microscope (SEM) and energy-dispersive X-ray spectroscopy (EDX) analyses for main morphology were performed on the fly ash samples. For the SEM/EDX analysis, a small sub-sample of the fly ash (<0.5 g) was placed directly on carbon tape. No further pre-treatment of the samples was made. The accelerating voltage of the SEM was 30 keV with large field detector (and X-ray cone). Different areas of the sample were investigated by SEM, and the element distribution was examined by element mapping using EDX on unpolished samples. Fly ash mineralogy was studied by X-ray powder diffraction (XRD), for identification of major crystalline phases. The instrument was a PANalytical X'Pert Pro operating at 45 mA and 40 kV applying Cu Kα radiation with a 2Θ X'Celerator detector. The samples were scanned in the range of 4-100 2 Θ within 8 hours. The diffractograms were interpreted by using the ICDD PDF-4 database for minerals.

3. Results and discussion

3.1. Morphology and mineralogy

To evaluate the similarity between the ashes particle size, major elements, mineralogy and morphology were investigated. Figure 1 shows the particle size distribution of all ashes, table 1 the major elements and mineralogy and Figure 2 shows the morphology of the fly ash particles.

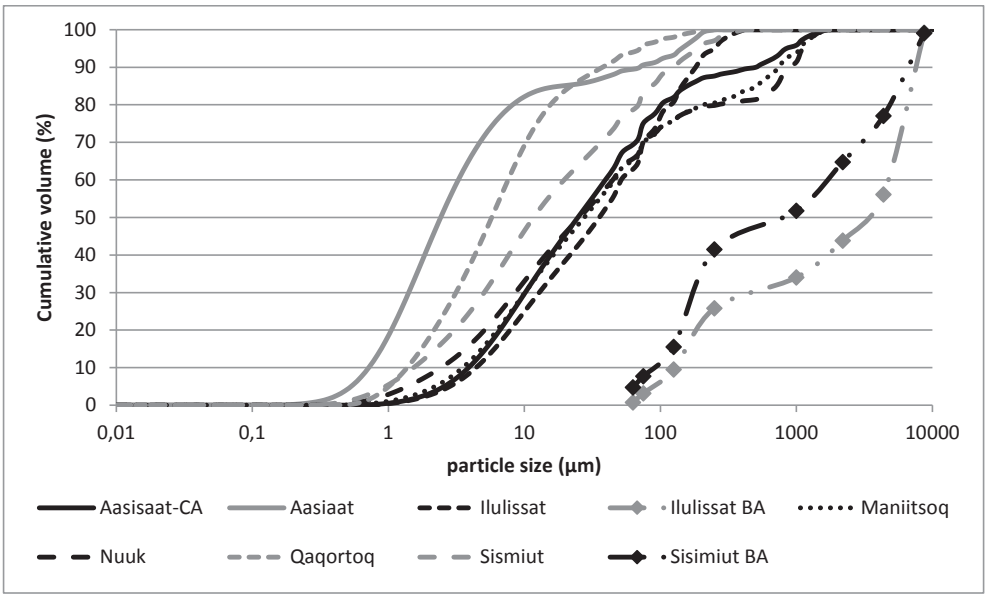

Figure 1: Particle size of fly ash, cyclone ash (CA) and bottom ash (BA)

The particle size distribution of the fly ashes varied greatly, however the main particles were less than 100 µm. Some similarities were seen in the particle size distribution between similar incineration plant types, especially for Maniitsoq, Ilulissat and Aasiaat – cyclone. For the Sismiut and Qaqortoq ashes, the Qaqortoq fly ash was finer than the Sisimiut fly ash. The bottom ashes were larger in particle size than the fly ash, as expected, and the Ilulissat bottom ash was larger in particle size than the Sisimiut bottom ash. The particle size of bottom ash is typically similar to soils used as road base material or structural fills and to produce the desired grain size distribution for these uses, bottom ashes are sometimes size processed (removal of parts of fractions or blended with other materials) [4]. Fly ash is more difficult to size process and possible uses should be based on the material as it is.

The elemental compositions of the fly ashes are presented in table 1. The amount of main oxides varied greatly between the ashes, although the main oxides were of Ca, Na, K, S and Si. A Cl content of 9 - 42 % in the ashes could hinder the reuse potential without pre-treatment due to corrosive and soluble properties of the fly ashes in the natural state. The main minerals identified were also connected to the elements found and indicates that Cl (NaCl and KCl) could

be easily removed by washing. The Aasiaat and Qaqortoq fly ashes revealed high LOI results, indicating a poor burn out of organic material, which most likely is due to insufficient temperature during the incineration, which could also imply formation of other unwanted contaminants, such as dioxins. Coal fly ash is widely used as supplementary cementitious material due to its pozzolanic and cementitious properties. According to DS/EN 450-1 for use of coal fly ash in concrete, the LOI should not exceed 9 %. A high LOI in cement will decrease the air content and give a more unstable concrete [5]. Cement consist mainly of CaO (approx. 65 %), SiO_2 (approx. 20 %) and Al, Fe and S oxides below 5 %, with the main minerals related to these compounds, called clinker minerals [5]. The composition of the fly ashes showed some similarities in the element composition with cement, however the Cl content should be removed and the pozzolanic properties be studied further.

Basic visual analysis of the fly ashes indicated two different main types, despite the three different incinerator types. Two fly ashes (Aasiaat and Qaqortoq) were black, very light almost electrostatic. The other fly ashes were greyish and denser. The SEM pictures (fig. 2) confirmed the particle size distribution in Figure 1 and revealed very fine-grained particles coating or covering larger spherical or rectangular particles. The fine-grained particles were likely to be polycrystalline platelets that accumulate in aggregates as seen by Eighmy et al. [6]. The Qaqortoq ash appeared to consist even more of these small aggregates than the Aasiaat ash. Both the Qaqortoq and Aasiaat ashes had a high water solubility (table 2) and a high content of water soluble elements as Na and K in the form of NaCl and KCl, which supports the lightness and porosity of these two ashes. SEM pictures of the other ashes show some similarities. The Nuuk and Sisimiut ash had similar appearance with both small and larger particles. The typical spherical fly ash particles usually found in coal fly ash were seen for the Maniitsoq, Ilulissat and Aasiaat ashes. The EDX analysis did not reveal the exact composition of these spherical particles, but they were likely a mix of Ti, Si, Mg and Al oxides. Generally, the elemental composition of the ashes was in the order of C>O>Ca>Cl for all the analysed ashes.

Table 1: Major elements and mineralogy for Greenlandic fly ashes

	Aasiaat-CA	Aasiaat	Ilulissat	Maniitsoq	Nuuk	Qaqortoq	Sisimiut
CaO (%)	42.0	12.6	42.0	46.2	42.0	15.4	25.2
SiO_2 (%)	10.1	3.9	10.3	9.2	6.2	2.8	4.9
Na_2O (%)	8.6	24.3	7.0	7.0	9.6	14.8	16.2
K_2O (%)	4.7	21.7	4.2	5.4	6.5	12.0	8.9
Al_2O_3 (%)	7.2	2.5	7.2	6.6	4.9	1.2	3.2
Fe_2O_3 (%)	1.6	0.6	1.9	1.2	0.8	0.7	1.3
MgO (%)	1.4	0.2	1.5	1.6	1.7	0.1	0.6
MnO (%)	0.1	0.0	0.1	0.1	0.1	0.1	0.0
P_2O_5 (%)	1.1	0.5	1.7	1.8	1.0	0.4	0.6
SO_3 (%)	4.7	3.0	3.7	5.2	6.7	2.2	3.0
TiO_2 (%)	1.7	1.0	2.5	2.0	1.4	0.6	0.9
LOI (%)	1.2	10.8	0.04	3	1.9	18.4	2.7
Cl (%)	11	42	8.9	9.8	13	27	23
Main minerals	KCl, NaCl, $CaCO_3$, Ca SO_4, SiO_2	KCl, NaCl, $CaCO_3$, SiO_2	KCl, NaCl, $CaCO_3$, Ca SO_4, SiO_2	KCl, NaCl, $CaCO_3$, Ca SO_4, SiO_2	KCl, NaCl, $CaCO_3$, Ca SO_4, SiO_2	KCl, NaCl, $CaCO_3$, SiO_2	KCl, NaCl, $CaCO_3$, Ca SO_4, SiO_2

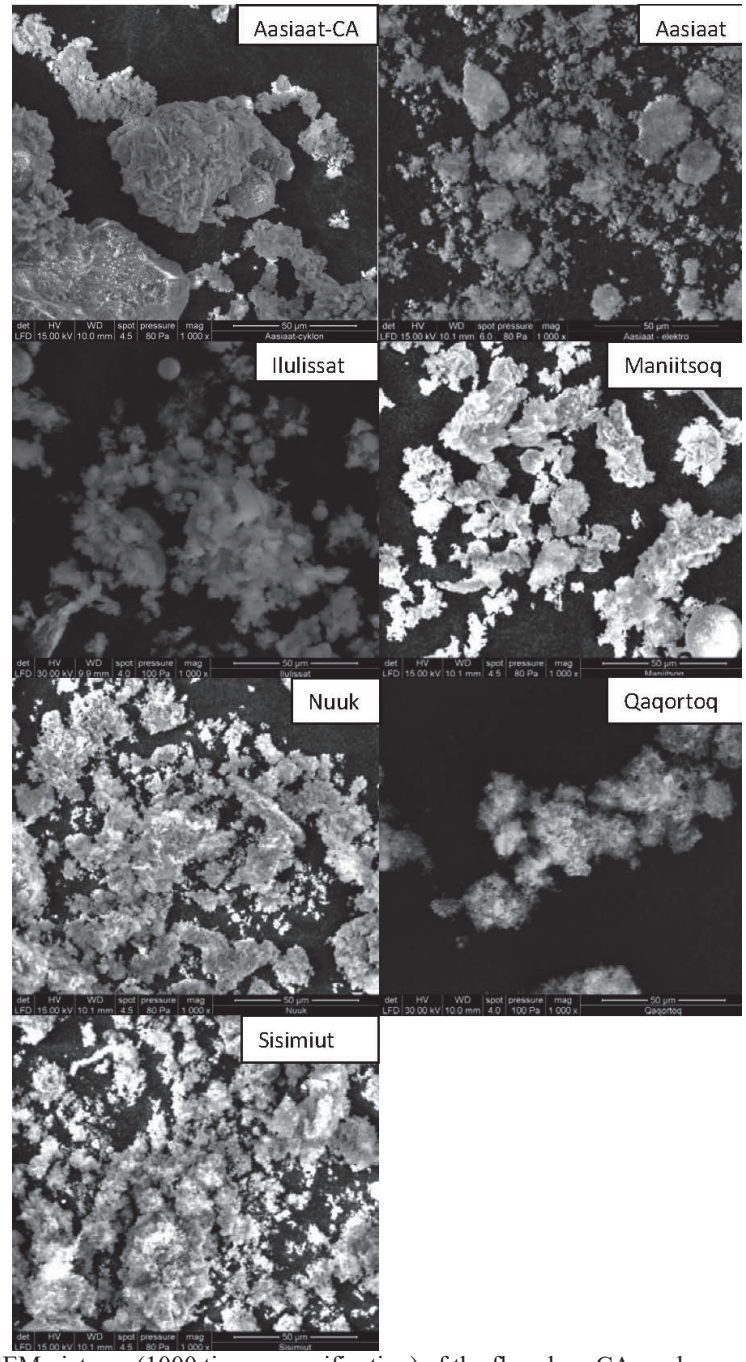

Figure 2: SEM pictures (1000 times magnification) of the fly ashes. CA-cyclone ash

3.2 Heavy metal content and availability

Table 2 shows the metal content and Table 3 shows the leaching of heavy metals from the incineration ashes. The incineration ashes are mainly alkaline; however the Aasiaat and Qaqortoq fly ashes are neutral in pH. The metal content also varied greatly in the ashes and similarities due to incinerator type was not seen. Nevertheless, Pb and Zn were found in the highest concentrations, which is usual for MSWI fly ash [7].

Table 2: Characteristics of incineration ashes. CA-Cyclone ash, BA-bottom ash

	Aasiaat-CA	Aasiaat	Ilulissat	Ilu-BA	Maniitsoq	Nuuk	Qaqortoq	Sisimiut	Sis-BA
pH	12.3	7.3	12.3	11.9	12.4	12.5	7.2	12.5	13.3
Water content (%)	1.5	1.5	0.8	n.a.	1.3	0.6	2.5	0.6	0.3
Water solubility (%)	28	69	22	n.a.	28	34	47	23	1.8
Al (g/kg)	28	6.8	59	47	24	28	8.7	125	27
As (mg/kg)	21	60	20	7.1	40	290	90	21	22
Ba (mg/kg)	883	292	985	518	365	380	249	334	1,120
Cd (mg/kg)	57	578	222	14	38	314	210	93	6.0
Cr (mg/kg)	221	111	304	136	188	165	115	70	72
Cu (mg/kg)	387	1,480	451	708	314	718	713	449	1,220
Mn (mg/kg)	682	196	979	371	383	284	339	403	360
Ni (mg/kg)	25	16	46	37	21	29	15	47	60
Pb (mg/kg)	1,000	6,270	2450	89	711	1,328	3,370	887	1,040
Zn (mg/kg)	12,440	27,780	12,720	2,270	7,250	13,220	36,600	6,850	4,300

The Qaqortoq and Aasiaat ashes were distinguished by a neutral pH and high water solubility. The high water solubility could not be directly related to the amount of leachable metals, as the metal leaching is mostly pH-dependent and not solubility dependent [7]. The bottom ashes contained high amounts of heavy metals of especially Cu, Pb and Zn and these metals also showed high deviation (not shown in table 2) due to metal pieces in the bottom ash. The heavy metal concentrations in the fly ash were less deviating.

Table 2: Metal leaching from the incineration ashes, CA-Cyclone ash, BA-bottom ash

	Aasiaat-CA	Aasiaat-BA	Ilulissat	Ilu-BA	Maniitsoq	Nuuk	Qaqortoq	Sisimiut	Sis-BA
pH	12.3	7.3	12.2	11.8	12.3	12.3	10.2	12.4	12.9
Al (µg/l)	<20	<20	38	275,000	287	178	793	466	240
As (µg/l)	<20	73	20	30	<20	32	88	1,240	<20
Ba (mg/l)	1.4	1.4	1.6	0.01	1.3	1.2	5.8	2.7	19.7
Cd (µg/l)	<20	<20	<20	<20	60	35	3,900	1,460	<20
Cr (mg/l)	6.4	3.9	8.9	2.0	16	5.4	1.6	12	0.01
Cu(µg/l)	82	2,330	24	36,700	253	300	60	74	40
Mn (µg/l)	<20	6,700	<20	<20	20	<20	27	<20	<20
Ni (µg/l)	<20	42	<20	90	191	89	36	<20	<20
Pb (mg/l)	30	394	45	0.02	19	37	2.1	0.5	2.5
Zn (mg/l)	4.0	1,450	3.2	0.14	5.8	5.5	3.6	1.1	1.2

No guideline levels exist for heavy metal leaching for reuse of fly ashes, but the environmental impact should be studied before using these materials as secondary materials. Using air pollution residues in mortar showed no significant difference in short term leaching compared to reference mortar [8], whereas using incineration residues in bricks increased the leaching of Cr [9]. Guideline levels for reuse in construction exist for bottom ash in Denmark [10], however the Ilulissat bottom ash exceeded the limits for Cr (< 0.5 mg/l), Cu (< 2000 µg/l) and Pb (< 0.1 mg/l) and the Sisimiut bottom ash exceeded the Pb limit.

4. Conclusion

- The fly ashes are characterised by small particles and high content of Ca, Na, K, S, Si and Cl. Ca, K and Cl are mainly present as NaCl and KCl which are highly soluble, which is also seen in water solubility of up to 70 % of the fly ash.
- All studied incineration residues contain high amounts of metals and also large fractions of the metals are leachable

- The incineration residues show potential for reuse, but pre-treatment may be necessary, such as removal of chloride and metals from fly ash and processing of particle sizes of bottom ash
- The Greenlandic waste incineration residue samples were different and evaluation of reuse should be taken individually, however the fly ash could have potential as cement replacement and bottom ash as sand replacement for construction purposes. Actual applications should be studied further.

5. Acknowledgements

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