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## MERCURY LEVELS IN FLY ASH AND APC RESIDUE FROM MUNICIPAL SOLID WASTE INCINERATION BEFORE AND AFTER ELECTRODIALYTIC REMEDICATION

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### ABSTRACT

Fly ash (FA) and Air Pollution Control (APC) residues collected from three municipal solid waste incinerators (MSWI) in Denmark and Greenland were treated by electrodynamic remediation at pilot scale for 8 to 10 h. The original residues and the treated material were analysed for mercury (Hg) in order to assess the influence of the electrodynamic treatment on the concentrations of this element. Mercury levels varied with the MSWI residue, ranging from 0.41 mg kg<sup>-1</sup> in FA sample from electrostatic precipitator (ESP) to 8.38 mg kg<sup>-1</sup> in MSWI residues from a semi-dry system with lime and activated carbon. Two distinct behaviours were observed for mercury as a result of the electrodynamic treatment. This element became enriched in the MSWI residues from the semi-dry system with activated carbon, whereas it decreased in ESP's and cyclone's FA. This work presents for the first time information about the effect of electrodynamic treatment on mercury levels and discusses the valorisation options for these MSWI residues.

*Keywords:* arctic, construction materials, EDR, electrodynamic remediation, Greenland, Hg, incineration, incinerators, MSW, MSWI, waste.

### 1 INTRODUCTION

In 2013, the Minamata Convention, an international treaty on mercury, was adopted worldwide in an attempt to protect the human health and the environment from anthropogenic emissions and releases of mercury. Mercury produces significant adverse neurological and other health effects, being particularly harmful to unborn children and infants [1]. The Convention identifies waste incineration as one relevant source for mercury, as listed in Annex D of Article 8 [1]. During waste incineration, mercury is released from a wide range of consumer waste products, such as batteries, paints, switches, electrical and electronic devices, thermometers, blood-pressure gauges, fluorescent and energy-saving lamps, pesticides, fungicides, medicines and cosmetics. For most of these products, mercury-free alternatives exist and are being adopted, resulting in less consumer products containing mercury. The exception is compact fluorescent light bulbs, which are being increasingly used [2].



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Mercury remains one of the most relevant global environmental issues, and stringent measures at EU level exist on emissions from different sources: 0.05 mg/Nm<sup>3</sup> [3]. To achieve emission limits, incinerators implemented air pollution control (APC) equipment, thus diverting mercury from the flue gas into municipal solid waste incineration (MSWI) residues. Fly ash (FA) and APC residue are generally fine powders, representing 4%–10% in weight of incinerated waste. In addition, APC residues are strongly alkaline, containing high concentrations of lime and other calcium compounds added for the cleaning process, as well as soluble metal chlorides [4, 5].

In the last 10 years, several researchers have extensively reported on the electrodynamic (ED) treatment of MSWI residues at lab and pilot scale [6–9]. These experiments targeted the removal of heavy metals and soluble salts from MSWI residues, aiming at the valorisation of this waste. Targeted heavy metals were mostly Cd, Cu, Cr, Pb and Zn. Two recent works have expanded the range of target pollutants to include previously unreported substances, such as As, Mn, Mo, Sb, Se, V, Cl and SO<sub>4</sub> [10] and also organic pollutants polychlorinated dibenzop-dioxins [11]. But so far, no report exists on the effect of the ED treatment on mercury content in MSWI residues, and this missing information is crucial to properly assess possible reuse options.

The objectives of this study were:

- To analyse the levels of mercury in the FA and APC residues from 3 MSW incinerators in Denmark and Greenland, equipped with different APC devices, and compare the results;
- To study for the first time possible changes in the content of mercury in different MSWI residues due to ED remediation;
- To discuss the implications of mercury levels for the reuse options of MSI residues.

## 2 MATERIALS AND METHODS

### 2.1 MSWI residues

Four different residues from municipal solid waste incineration (MSWI) were used in this work (Fig. 1):

**FA:** FA collected from the electrostatic precipitator (ESP) in a MSWI facility in Nuuk (Greenland), without addition of any substances for APC. The facility is a small incineration plant with an installed capacity of 10,000 t year<sup>-1</sup> of municipal solid waste, working at comparatively low temperatures.



Figure 1: FA and air-pollution control (APC) residues from the incineration of municipal solid wastes: **FA** (FA from the ESP of an incinerator in Greenland), **WET** (APC residue from a wet-system in Denmark), **SD** (APC residues from a semi-dry system), **SD carb** (carbonated SD sample).

**WET:** FA collected from bag filters and cyclone (prior to the wet scrubber) at plant Vest-forbrænding, Glostrup (Denmark). Activated carbon is added to the fabric filters (FF) to remove dioxins and Hg from the flue gas.  $\text{NH}_3$  is injected into the furnace to reduce emissions of  $\text{NO}_x$ . The facility is a large-scale incinerator with state-of-the-art APC equipment, and an installed capacity of 600,000 t of municipal solid waste per year.

**SD:** A mixture of FA and flue gas cleaning residues from the MSWI plant REFA I/S in Nykøbing Falster (Denmark) collected by ESP after a semidry treatment process consisting of slaked lime and activated carbon in the injection.  $\text{NO}_x$  is removed by selective non-catalytic reduction. The facility is a large-scale incinerator with state-of-the-art APC equipment and an installed capacity of 177,000 t year<sup>-1</sup>.

**SD carb:** Sample obtained from SD by inducing carbonation. For this purpose, a sub-sample of SD was kept at room temperature under moist conditions for several weeks. SD residues are lighter in colour than the other residues, likely due to the added lime. Carbonation did not change the appearance of the material (colour or physical form) when compared to the original non-carbonated sample.

In order to avoid clogging of the ED apparatus, the fraction >1 mm of the MSWI residues was dry sieved out and only the fraction <1mm representing 99% of the mass was used in the experiments.

## 2.2 Electrodealytic pilot plant

The pilot plant consisted of an ED stack which was continuously fed the APC/FA suspension (diluate), using pumps. Inside the ED stack, ions dissolved from the ash were transported by electromigration through ion-exchange membranes into the concentrate flow (Fig. 2).

The concentrate consisted of 30 L tap water with 400 mL 1:1  $\text{HNO}_3$ , and the electrode of 30 L distilled water with 400 mL 1:1  $\text{HNO}_3$ . The ED stack consisted of 34 or 50 cell pairs (530 cm<sup>2</sup> active surface area per cell) with Neosepta cation and anion exchange membranes. The spacers were 0.6 mm and 5 mm thick for the concentrate and diluate, respectively. There were electrode compartments in each end of the ED stack with electrode clean solution circulating between them. The electrodes were titanium plates covered by a layer of metal

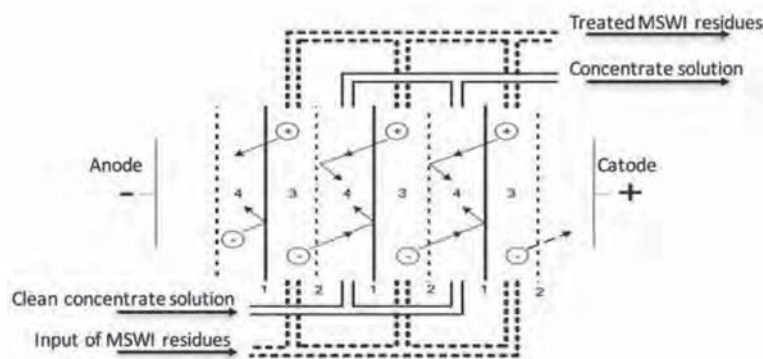


Figure 2: Principle of electrodealytic treatment. 1 and 2 - ion exchange membranes; 3 - APC residue in suspension (diluate) compartments; 4 - concentrate compartments.

Table 1: Experimental conditions for the electrodialytic experiments.

Experiment number	1	2	3	4	5	6
MSWI residue	FA	WET	SD	SD	SD	SD carb
Cell pairs (number)	5	34	50	34	34	50
Time (h)	10	8	10	10	10	10
Residence time (h)	1.8	1.4	1.8	1.7	1.7	1.8
Current (mA/cm <sup>2</sup> )	4.7	5.7	5.7	5.7	5.7	5.7

oxides. The flow rates were adjusted in each experiment to maintain overpressure in the diluate compared to the concentrate, with the diluate flow rate always kept at a maximum. The electrode clean solution tank was ventilated for exhaust gases such as chlorine from the electrode reactions. A more thorough description of the pilot plant can be found in Kirkelund *et al.* [12]

### 2.3 Electrodialytic experiments

Six ED experiments were made in total, varying the MSWI residue: FA (experiment 1); WET (experiment 1); SD (experiments 3–5); and SD carb (experiment 6). The experimental conditions are shown in Table 1. L/S was kept constant at 10 L/kg in all experiments, with either 8 kg MSWI residue and 80 L tap water in experiments 3 and 6, 5.3 kg MSWI residue and 53 L tap water in experiments 2, 4 and 5, and 1 kg of MSWI residue and 10 L tap water in experiment 1, dependent on the number of cell pair which results in different volume of the stack. At the end of the ED experiments the diluate was filtered through filter paper at normal pressure, and the MSWI residue remaining was dried in an oven at 40°C prior to further analysis.

### 2.4 Leaching experiments

Leaching of Hg from the MSWI residues was assessed before and after the ED experiments according to DS/EN 12457-1 in batch [13]. The test was carried out at L/S 2 (L/kg) by mixing 10 g of MSWI residue with 20 mL deionised water. The suspension was shaken for 23 h on an end-over-shaker. The pH of the suspension was measured before vacuum filtration through a 0.45 µm nucleio filter. The filtrate was analysed for total Hg.

### 2.5 Analytical

Total Hg content was analysed according to Danish Standard DS259 [14] where 1 g of MSWI residue and 20 mL 7.3 M HNO<sub>3</sub> was heated at 200 kPa (120°C) for 30 min. The liquid was thereafter separated by vacuum filtration through a 0.45 µm filter and diluted to 100 mL. All Hg measurements were made by atomic absorption spectroscopy with hydride generation (HGAAS).

### 3 RESULTS AND DISCUSSION

#### 3.1 Hg in MSWI residues

Total concentration of mercury in the four MSWI residues used in this work ranged from 0.41 to 8.4 mg kg<sup>-1</sup> (Table 2). The WET and SD samples came from modern incinerators in Denmark using activated carbon and limestone slurry. It has been found that activated carbon adsorbs large amounts of both Hg<sup>0</sup> and Hg<sup>2+</sup> and that limestone slurry adsorbs large amounts of Hg<sup>2+</sup> [15]. It follows that systems using activated carbon and slaked limes, such as our samples WET and SD, are more effective in capturing Hg, so higher levels are found in the collected residues, and only a minor fraction of mercury is released to the atmosphere.

Comparing mercury levels in the samples before and after the carbonation (SD vs SD carb) showed a reduction of 44% (Table 2). During carbonation, the carbon dioxide from the atmosphere reacts with the alkaline material in the residue, leading to a pH decrease and to the precipitation of calcite (a process that can take from several days to several thousand years). The carbonation phenomenon increases the total mass of the residue, where bound CO<sub>2</sub> may amount to 25%–50% of the initial mass of the residue [16]. In practice, this translates into a dilution effect, which explains the decreased mercury concentration in the carbonated sample, SD carb, when compared to the non-carbonated SD. A similar behaviour was previously found for dioxin levels [16], in which dioxin levels in carbonated APC residues decreased between 5% and 53% when compared to similar, but non-carbonated material.

Hg levels in FA from the Nuuk incinerator in Greenland (FA) were 20 times lower than MSWI residues collected from the incinerators in Denmark, likely due to the poor retention capacity of mercury in ESPs. According to Zhang *et al.* [15]), particulate-bound mercury accounts for less than 5% of mercury in flue gas. ESP are designed to capture solid particles, not vapours, so the Nuuk incinerator is possibly capturing only a small fraction of total mercury in the flue gas, which is the fraction adsorbed onto the surface of the FA. As no other air pollution cleaning devices are used in the Nuuk incinerator, the remaining mercury, representing between 85% (17) and above 95% of total mercury content in the waste, possibly gets released to the atmosphere.

The impact of these emissions depends on the mercury speciation profiles in the emitted gas. In combustion processes, including waste incineration, approximately half the mercury is emitted as gaseous elemental mercury (Hg<sup>0</sup>) and half as gaseous oxidised mercury (Hg<sup>2+</sup>) [17]. Residence time in the atmosphere for elemental mercury is 0.5–1 years [17]), whereas Hg<sup>2+</sup> is more soluble and can be washed out in water droplets and adsorbed to surfaces, thus having a shorter residence time, in the order of hours to days ([15]). This is important because

Table 2: Concentration of mercury (mg kg<sup>-1</sup>) in MSWI residues (\*average and standard deviation of two samples; \*\*average and standard deviation of four samples).

	FA	WET	SD	SD carb
Hg (mg kg <sup>-1</sup> )	0.41	7.35±3.7*	8.4±2.3**	3.7
System to control flue gas emissions	ESP	fabric filters and cyclones with addition of activated carbon	Injection of slaked lime and activated carbon, followed by ESP	

emitted  $\text{Hg}^{2+}$  is deposited regionally, increasing local mercury levels [17], while elemental mercury remains in the atmosphere long enough to be transported around the globe.

Following wet or dry deposition, inorganic mercury reaches aquatic systems, being further transformed into methylmercury by bacterial action. Methylmercury is toxic and biomagnifies as it moves up the food web. Consumption of fish and other marine foods can increase the risk of human exposure, especially among indigenous populations with diets based on marine foods ([2]). This is the case for Greenlandic people, who consume marine predators at the top of the marine food web, such as fish, seals and whales.

Total mass emissions of mercury depend on mercury levels on the waste, on the efficiency of APC system for scavenging mercury from the flue gas, and also on the total amount of waste incinerated. Emission factors in modern Danish incinerators are  $3.4 \times 10^{-2}$  g Hg emitted/g Hg in waste, while in smaller and older Greenland incinerators emission factors are 25 times higher, approximately 0.85 g Hg emitted/g Hg in the waste [18]. Considering that the common goods in Greenland in general are similar to comparable goods in Denmark, the mercury content of waste is assumed to be equal [18]. On the other hand, the amount of incinerated waste is much lower for the Nuuk incinerator ( $9\,371 \text{ t year}^{-1}$ ) than for any of the Danish ones (I/S Vestforbrænding:  $522\,258 \text{ t year}^{-1}$ ; I/S REFA  $111\,039 \text{ t year}^{-1}$ ), meaning that the Nuuk facility emits in total less mercury per year than the larger plant I/S Vestforbrænding. However, the local impacts of Hg emissions from combustion sources must be viewed against the fragile arctic environment and the biomagnification of Hg in this environment, together with the high dependence of the Greenlandic diet on marine food consumption. Eisted and Christensen [18] have modelled the impact of air emissions from the small MSW incinerators in Greenland using LCA approaches, and reported a high impact on human toxicity, especially from mercury emissions. The Hg emissions and possible impacts from local sources such as Greenlandic waste incineration plants is not fully understood and should be investigated and compared to the impact of globally emitted Hg that accumulate in the Arctic.

### 3.2 Effect of Electrodialytic Treatment on Mercury Concentrations in FA and APC Residue

Six ED experiments were made in total, varying the MSWI residue. Hg levels in the solid materials before and after ED treatment are shown in Fig. 3a. A decrease of mercury concentration as a result of the ED treatment was observed in samples FA and WET, indicating that during treatment mercury was dissolved from the MSWI residue and transported into the concentrate chambers, as expected. On the opposite, in APC residues (SD and SD carb) the mercury concentrations increased after ED treatment, with SD carb samples presenting more than three times the initial concentration. The increase is likely due to the MSWI residue partially dissolving during treatment, while mercury becomes enriched in the non-dissolved fraction, similarly to what was seen for dioxins in other studies [11]. This is probably enhanced by the activated carbon in the APC residues, to which mercury strongly adsorbs.

Figure 3b shows leaching values obtained. For FA and SD carb the determination was not carried out after ED treatment, so only the results of the original residue are shown. It can be seen that ED treatment resulted in increased leaching for the WET sample and also for two batches of SD samples (exp.3 and exp.4). The SD batch used in experiment 5 had different properties than those used in experiment 3 and 4, characterised by a much higher initial mercury level, likely due to variations in input materials or in operational conditions at the

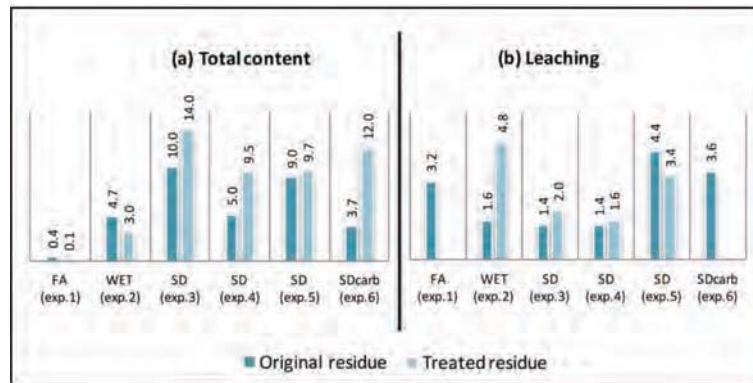


Figure 3: Comparison of mercury total levels and leaching characteristics of MSWI residues before and after ED experiments: (a) mercury concentration (mg/kg dry matter); (b) leached mercury (µg/kg dry matter).

incinerator. SD in experiment 5 was the only sample where leaching was decreased after ED treatment, among those assessed.

### 3.3 National regulations and target values

Aiming at waste minimisation and resource conservation several possible reuse/recycling options for MSWIFA have been identified in the last two decades, namely cement production, geotechnical applications (e.g. road construction), glass and glass ceramics, agriculture, use as stabilising agent, use as adsorbent and for zeolite production [19,20]. In assessing these options, the levels of heavy metals and soluble salts have been extensively considered previously, whereas mercury content has barely been discussed.

Under current legislation MSWI FA and APC residues are considered hazardous waste and their use is prohibited in general. Therefore, limit values for acceptable contents and leaching of toxic constituents set for the use of residual products in geotechnical application [21] are not valid for these materials. Nevertheless, the values set in [21] are considered to be the most appropriate legislative limits to consider when discussing the possibility of using upgraded MSWI APC residues in geotechnical applications in Denmark, such as road construction. Three different categories are possible: **category 1** (unrestricted use) is defined by a low total content of contaminants and water leachability; **category 2** (restricted use) and **category 3** (severely restricted use) are based on water leachability of the contaminants at a liquid to solid ration (L/S) equal to 2. The limits are shown in Table 3. From this table and from Fig. 3 it can be seen that:

- The original FA residue falls within category 1 (unrestricted use) regarding total mercury content, but then fails to meet the leaching limit for all three classes.
- Original WET sample meets leaching criteria for class 3 (severely restricted) use, but after ED treatment leaching increases, and the sample no longer complies;
- SD batches used in experiments 3 and 4 comply with leaching criterion for mercury for class 3 (severely restricted) use, both before and after treatment. However, SD batch used in experiment 5 exceeds this criterion.

Table 3: Mercury legislative limits for total content and water leaching (batch tests at L/S 2 L/kg with distilled water) in residues for geotechnical applications in Denmark, according to [22] (leaching values recalculated from ug/L in the eluate to  $\mu\text{g/kg d.w.}$ ).

Category	1 unrestricted use	2 restricted use	3 severely restricted
Hg total content ( $\text{mg kg}^{-1}$ )	1	-	-
Hg leaching ( $\mu\text{g kg}^{-1}$ )	0.2	0.2	2.0

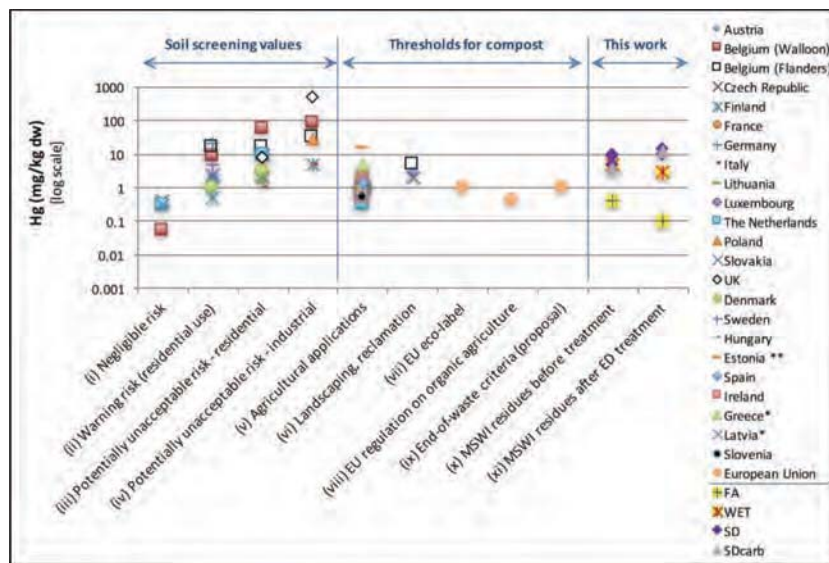


Figure 4: Limit values for mercury in different materials (\* regulations referring to waste management; \*\* regulations referring to sludge).

It is also of interest to know how the concentrations of Hg in the MSWI residues in this work compare against some of the regulatory limits for compost and digestates across different nations and also against screening values for soils. A tentative comparison is shown in Fig. 4.

On the right side of Fig. 4 are displayed the values obtained in the current work for MSWI residues, before and after ED treatment (x–xi). On the left side of figure the screening values for soils across several nations are displayed, compiled from Carlon [23], and grouped into 4 categories: (i) negligible risk, (ii) warning risk for residential sites, (iii) potentially unacceptable risk for residential sites, and (iv) potentially unacceptable risk for industrial sites. Screening values are lowest for negligible risk and highest for unacceptable risk for industrial sites. The screening values within each category have a high variability, with the highest and lowest values differing around a factor of two orders of magnitude. Of the MSWI residues used in this work, only FA could be framed within negligible risk category in what relates to mercury levels, whereas the other 3 residues would fit into one of the other three categories, depending on the country.

The middle section of Fig. 4 displays existing limits for mercury in (v) compost for agricultural applications, compiled from [21] (vi) compost and digestates for non-agricultural applications, such as landscaping and reclamation, compiled from [21]; (vii) EU eco-label limits for soil improvers and growing media, according to 2007/64/EC and 2006/799/EC; (viii) EU regulation on organic agriculture - compost from source separated biowaste; and (ix) End-of-Waste criteria (proposal) for compost and digestates [21].

From the graph it can be seen the highest and lowest value for mercury levels in the middle section differ around one order of magnitude, being lower for these agriculture-related applications than the screening values for soils (except for negligible soil risk). Only treated FA is below the maximum acceptable levels of mercury in compost for agricultural and non-agricultural application. The remaining residues used in this work are all above maximum acceptable levels, and differences between treated and untreated residues are not relevant enough to change the relative position of MSWI residues in the graph.

#### 4 CONCLUSION

Differences in the Hg levels in the three MSW incinerator residues analysed were attributed to technological differences. The Nuuk incinerator is an older, small-scale incineration facility where the only flue gas treatment is the ESP and where activated carbon is not used, resulting in a poor retention of mercury and higher emissions to the atmosphere. Even though the amount of incinerated waste is low, the low level of emission control might be relevant against the fragile Arctic environment, given the dependence of Greenlandic people on marine food consumption.

ED treatment generally increases both total levels and leachability of mercury in MSWI residues, although some deviations to this behaviour were observed. Generally speaking, both original as well as treated residues have mercury levels and leachability that prevents their possible application either for road construction or for agricultural and soil related applications (land reclamation, landscaping). For other applications it was not possible to find national regulations or target values that would allow any comparison.

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