

# Mass-balance estimation of heavy metals and selected anions at a landfill receiving MSWI bottom ash and mixed construction wastes

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## Abstract

An estimation of the heavy metal and anion mass-balance was made for municipal solid waste incinerator bottom ash deposited at a construction and industrial waste landfill. The mass-balance was found by comparing the content of metals and anions in the landfill leachate to the metal and anion content in the deposited bottom ash. The discharge of heavy metals ranged from 0.001% for Pb to 0.55% for Cr, which is approximately at the same level as in regular municipal solid waste (MSW) landfills. Landfilled organic material and silicates from construction waste might have contributed to the retention of metals. Chloride, and to a lesser extent sulphate, appeared to be readily released from the landfill. It was estimated that a mass corresponding to 80% of the  $\text{Cl}^-$  and 18% of the  $\text{SO}_4^{2-}$  in the bottom ash was discharged annually. Low retention, especially of chloride, may lead to a rapid decline in the discharge of this ion in the future when the landfilling of bottom ash is discontinued.

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## 1. Introduction

The limited availability of land for landfills and the possibility of utilization of the energy in the waste are two of the major reasons for incinerating municipal solid waste (MSW) instead of landfilling it. Incineration offers a reduction of around 70% of the mass and 90% of the volume of MSW [1].

Due to increasing restrictions on the emission of pollutants to the environment, MSW incinerator (MSWI) plants in operation have been subject to continuous technical upgrading. A remaining problem is the handling of residues after incineration. Heavy metals from MSW are pre-concentrated in bottom ash and particularly in fly ash [2]. Fly ash collected from flue-gas is generally regarded as hazardous waste, and handled accordingly. Bottom ash is usually landfilled [3], but has also been utilized for road construction and similar

purposes [4]. Bottom ash from Norwegian MSW incinerator plants is usually dealt with by depositing it at MSW landfills or at landfills where construction and inorganic waste unsuitable for incineration is deposited.

The concentration of most heavy metals in MSWI bottom ash is higher than in general MSW. In a mass-balance investigation performed by Heie and Sørum [2], the metal content in general MSW and MSWI bottom ash was determined. The concentration of Cd, Cr, Fe, Pb and Zn was 3–4 times higher in the bottom ash compared to the MSW prior to incineration (Table 2). It is often assumed that metals are discharged from a MSWI bottom ash landfill at a higher rate than MSW landfills.

This paper describes an investigation of the mass-balance of selected metals and anions in MSWI bottom ash deposited at a landfill where the ash is known to influence the leachate to a significant degree. The metal mass-balance estimation is compared with similar estimations for MSW landfills. The estimation was done by calculating the total mass of metals

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and anions deposited through the bottom ash, and comparing this to the total discharged mass of the same species in the leachate. It is vital to assess the mass-balance in order to determine the fate of heavy metals and anions contained in landfilled bottom ash, and the potential effects of co-deposition of MSWI bottom ash and other wastes.

## 2. Materials and methods

### 2.1. Waste incinerator plant

The bottom ash from the Bergen Interkommunale Renovasjonsverk (BIR) MSWI plant in Bergen, Norway, was chosen for the investigation. This is a Von Roll fluidised bed incinerator plant that began operation in 1999. In 2002 and 2003, a total of about 90,000 t MSW was incinerated annually. A portion of the paper, glass and metals were source-separated from the waste stream before incineration.

An electrostatic precipitator along with wet and dry scrubbers was installed to purify the flue-gas. The bottom ash and the fly ash were handled separately. The bottom ash was continuously collected, and magnetic metals extracted from the ash. The remaining bottom ash was landfilled at the Mjelstad Miljø landfill at Osterøy, Norway.

### 2.2. Description of landfill

The landfill was established in 1999 in a narrow valley in Osterøy, Norway, an area with an annual precipitation of 2160 mm. By the end of 2003, an area of approximately 11,000 m<sup>3</sup> was filled up to a height of 20 m. This contained approximately 300,000 t of waste, of which 52,000 t were deposited in 2002 and 51,000 t in 2003. No wet organic waste or MSW had been deposited. The landfill was lined with an impermeable polyethylene liner in addition to a clay geomembrane, but a top liner was not established. Both surface- and ground water around the site was sampled and analysed four times a year, and no leakages in the liners were observed. The leachate was collected at the bottom of the landfill, and discharged through a separate pipeline.

### 2.3. Sampling and analysis of bottom ash and landfill leachate

Batches of 3–7 kg bottom ash were collected by “stopped-belt” sampling. Eight samples were collected in 2002 and 12 in 2003. The samples for each year were homogenized and mixed, and representative gross samples acquired from the material. In both 2002 and 2003, the total mass of bottom ash sampled was approximately 60 kg.

The volume of the discharged leachate was continuously measured, and samples were collected as homogenized volume proportional samples. At least 1.5 l of leachate was sampled each time. The pH of the leachate was measured in field with a Hanna Insitu 9026/HI 9023 pH instrument.

All samples were stored dark in polyethylene containers, and sent for analysis within 24 h after sampling.

Both the bottom ash and the homogenized unfiltered leachate were acid digested prior to metal analysis. Analysis of Pb, Cr and Cd was done by GF AAS (Perkin-Elmer Analyst 800) in 2002, and ICP-MS (Agilent 7500c) in 2003. Other metals were determined by ICP-OES (Perkin-Elmer Optima 3000 DV). Anions in the bottom ash were extracted into solution by shaking dried bottom ash in deionised water (L/S = 20) for 2 h, with subsequent filtration. The anion level determined thus represented only the water-soluble part of the total. Anion determination in both extract and leachate samples were done by ion chromatography (Dionex DX-10 ion in 2002, Lachat QuikChem 8500 ion in 2003, both with a Dionex AS-14 column).

## 3. Results and discussion

### 3.1. Slag and ash generated from the incinerator plant

The masses of the MSWI residues after incineration are summarized in Table 1.

In 2003, the portion of the bottom ash with particle diameter larger than 10 mm was separated from the rest by use of a grate drum. This part, amounting to  $4146 \times 10^3$  kg, was stored while awaiting later metal recovery. The removal of particles larger than 10 mm was not expected to alter the level of heavy metals in the landfilled bottom ash, since these are mainly present in particulate matter smaller than 10 mm [5].

The bottom ash residue amounted to approximately  $18,000 \times 10^3$  kg in 2002 and  $20,000 \times 10^3$  kg in 2003. Only the portion of the bottom ash left over after magnetic removal of iron (2002), and magnetic removal/sieving (2003), was deposited at the landfill. The magnetic removal of iron resulted in a 10–14% reduction of the bottom ash mass.

### 3.2. Chemical composition of the bottom ash

The concentrations of metals and anions in the landfilled portion bottom ash were determined. These data are presented in Table 2, along with literature values for MSW and bottom ash from an investigation at a Norwegian MSW incinerator plant during 1996 [2].

Table 1  
Mass of MSW incinerated and the resulting residue generated at the BIR incinerator plant in year 2002 and 2003

Waste/residue fraction	Generated masses (10 <sup>3</sup> kg)	
	Year 2002	Year 2003
Total incinerated MSW	88500	91060
Fly ash	1838	2466
Bottom ash to landfill	16160	14450 <sup>a</sup>
Magnetic metal recovery	2261	1551
Bottom ash >10 mm	–	4146

<sup>a</sup> In 2003, only bottom ash <10 mm in particle size was landfilled.

Table 2

Level of selected metals and anions ( $\text{mg kg}^{-1}$ ) in bottom ash sampled at the incinerator plant during 2002 and 2003 (all results reported on wet weight ash (82 and 81% dry matter in 2002 and 2003, respectively)), and literature values of metals in MSW and in bottom ash (wet weight) after incineration of the MSW [2]

Parameter	Literature values		Bottom ash sampled and analysed in this study		
	MSW prior to incineration	MSWI bottom ash	Landfilled bottom ash (2002)	Landfilled bottom ash (2003)	Average (2002 and 2003)
Cd	5.4	15	11	2	6.6
Cr	21	100	100	130	120
Cu	n.d.	n.d.	2700	1500	2100
Ni	n.d.	n.d.	110	50	80
Zn	1100	4400	4200	6600	5400
Hg	1.5	0.50	0.12	1	0.56
Pb	450	2000	1300	2400	1800
Chloride	n.d.	n.d.	2500	3400	3000
Sulphate	n.d.	n.d.	1100	3200	2200

n.d.: not determined.

Cd, Hg and, to some extent, Pb and Zn accumulate in the ash and gas-scrubbing residue, with lower concentrations in the bottom ash [3,4,6]. The refractory elements such as Cr and Cu, however, are predominantly present in bottom ash [4].

### 3.3. Quality of leachate from the bottom ash landfill

Results from the analysis of the landfill leachate throughout 2002 and 2003 are shown in Table 3. Some of the results have relatively high standard deviations, probably due to high rainfall prior to one sampling occasion (December 2003). High rainfall can affect the leachate composition due to changes in water flow-paths in the landfill during such episodes [7]. Table 3 also includes average concentrations in leachate from four MSW landfills in the same geographical area as the MSWI bottom ash landfill. The four MSW landfills have been described in more detail by Øygard et al. [8].

Table 3

Concentration of selected variables in the leachate from the MSWI bottom ash landfill, 2002–2003

Variable	Mean concentration for MSWI bottom ash landfill	Range of mean concentrations for MSW landfills in the same area
pH	$7.8 \pm 0.5$	6.6–7.2
$\text{COD}_{\text{Cr}}$ ( $\text{mg L}^{-1}$ )	$2400 \pm 1100$	190–2500
Chloride ( $\text{mg L}^{-1}$ )	$1900 \pm 380$	56–590
Sulphate ( $\text{mg L}^{-1}$ )	$554 \pm 193$	9.8–21
Fe ( $\text{mg L}^{-1}$ ) <sup>a</sup>	$1.3 \pm 1.4$	34–150
Cd ( $\mu\text{g L}^{-1}$ ) <sup>a</sup>	$1.2 \pm 2.8$	0.12–0.31
Pb ( $\mu\text{g L}^{-1}$ ) <sup>a</sup>	$19.0 \pm 31$	1.8–6.3
Hg ( $\mu\text{g L}^{-1}$ )	$0.26 \pm 0.25$	0.01–0.02
Cr ( $\mu\text{g L}^{-1}$ )	$500 \pm 310$	9.6–56
Cu ( $\mu\text{g L}^{-1}$ )	$290 \pm 270$	3.3–26
Ni ( $\mu\text{g L}^{-1}$ )	$64 \pm 32$	7.4–25
Zn ( $\mu\text{g L}^{-1}$ )	$58 \pm 36$	55–410

Mean and S.D. given. For Cr, Cu, Ni and Zn,  $n=4$ , otherwise  $n=8$ . Ranges of mean concentrations (2002–2003) for leachate from four MSW landfills in the same geographical areas as the MSWI bottom ash landfill.

<sup>a</sup> The high standard deviation is caused by unusually high concentrations in the sample in December 2003.

Leachate concentrations of chloride and especially sulphate were many times higher than those in leachate from MSW landfills in the same geographical area. This may be due to high levels of mobile  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  in the bottom ash. Conversely, the concentration of Fe was low in comparison to that from MSW landfills. Of the heavy metals, concentrations of Cu, Cr and Hg were higher in the leachate from the bottom ash landfill compared to the MSW landfills.

### 3.4. Mass-balance estimation

An estimation of the mass-balance of heavy metals and selected anions in the MSWI bottom ash deposited at the landfill was made using the average metal and anion content in the generated bottom ash (Table 2) and the average metal and anion content in the leachate (Table 3). Since the landfill liners isolate the leachate from the surrounding environment, metals and anions are only discharged through the leachate sampling point. Evaporation of Hg has not been taken into consideration.

The estimation does not account for all the metals or anions in the landfill, since the waste deposited from the initial landfill start-up until the end of 2001 has not been included on the input side of the mass-balance. In addition, the deposited ash accounted for only 30% of the total mass of waste deposited at the landfill through 2002–2003. A further 12% was waste foundry sand. The remaining 58% was construction waste, which included cardboard, wood and plasterboard (a potential source of sulphate). In a Danish investigation, plasterboards were found to represent on average 3.1% of the mass of deposited construction waste [9].

The MSWI bottom ash was assumed to be the main source of metals and chloride, with only low levels originating from the construction waste and foundry sand. The black colour and the high level of chloride in the leachate were indicative of the influence of the bottom ash. Although the other waste was assumed not to contribute significantly to the amount of metals and chloride in the landfill, it probably influenced the rate of leaching of metals. In particular, adsorption sites on organic material probably contributed to the metal retention.

Table 4

Total discharges of selected elements/ions through the landfill leachate during 2002 and 2003, and the deposited mass of the elements/ions in the same period of time

Variable	Total mass deposited through bottom ash (kg)	Total mass detected in leachate (kg) <sup>a</sup>	Total discharge that might be caused by leaching of bottom ash (%)
Chloride	90000	72000	80
Sulphate	66000	21300	32
Cd	200	0.048	0.024
Pb	57000	0.72	0.001
Hg	17.0	0.010	0.059
Cr	3500	19.0	0.55
Cu	64000	11.0	0.017
Ni	2500	2.5	0.10
Zn	165000	2.2	0.001

<sup>a</sup> Calculated from the measured leachate volume during 2002–2003, and average element/ion values measured in leachate during the same time.

Thus, the rate of leaching of metals from MSWI bottom ash monofills might be higher than values presented in Table 4.

#### 3.4.1. Mobility of heavy metals

Only a small percentage of the heavy metals landfilled through the bottom ash during 2002–2003 were leached from the landfill during this period. Such high retention was predicted by Stegemann et al. [1].

For Pb, the discharge in leachate equalled 0.001% of that deposited through bottom ash, a discharge rate similar, if not lower than that reported for MSW landfills [8], and corresponding well to values reported by Belevi and Baccini [10]. For Hg and Cd, the estimated discharges of 0.06 and 0.02%, respectively, are similar to values for MSW landfills in the same area [8].

Chromium had the highest discharge of the heavy metals studied, both in terms of total mass (19 kg) and percentage deposited through ash (0.55%). The concentration in the leachate is higher than found in the MSW landfills. The percentage discharge of this metal is lower, however, than that found for MSW landfills (0.60–1.04%) [8]. Since Cr is precipitated as hydroxides rather than sulphides, it is likely that this metal would behave similarly in a MSWI bottom ash compared to a regular MSW landfill.

**3.4.1.1. Cause of metal retention.** Retention of heavy metals in regular MSW landfills is usually related to sulphide precipitation at reducing conditions [11], or binding to humic substances [12]. Due to the presence of both organic matter and sulphur, it is likely that sulphide precipitation occurs also in this landfill. There does not, however, appear to be sufficient reduction potential in the landfill to reduce all the sulphate to sulphide, which otherwise would lead to an almost complete retention of sulphate along with metals [12].

The presence of carbonates was confirmed by the visual observation of gas development when acidifying the leachate. Although the solubility of metal hydroxides and carbonates are generally higher than the corresponding sulphides, many have a low solubility at the slightly basic pH (pH 7.8) of the leachate. Formation of metal carbonates and hydroxides thus plays a significant role in metal immobilization in the special

case of landfills for bottom ash [7,13–15]. Generally, a rising pH in the landfill/leachate leads to an increased retention of metals [10]. However, the solubility of Zn- and Pb-hydroxide increases at elevated alkaline pH, due to their amphoteric nature [16,17].

As pointed out by Johnson et al. [14], the solubility of heavy metals is not controlled solely by precipitation as sulphides, carbonates and hydroxides. Binding/adsorption to humus/solid matter from cardboard/wood could be considerable, particularly for Zn, Pb and Cu [13,18]. In a true MSWI bottom ash monofill containing lower levels of organic matter, this retention would be much less. Oxidized aluminium compounds and silicates, abundant in MSWI bottom ash, are also potent adsorbents of metals [6,13,16].

The concentration of Fe in the leachate is low compared to the concentrations found in leachate from local MSW landfills (Table 3). This may be due to magnetic removal of Fe from the ash prior to landfilling. However, additional Fe from the deposited construction waste has not been accounted for. Alternatively, the retention might be higher than at the comparable MSW landfills, due to the lower solubility of Fe at the higher pH found here.

#### 3.4.2. Anions

The discharge of  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  through the landfill leachate amounted to approximately 80% of the chloride and 32% of the sulphate deposited as bottom ash. However, the mixed construction waste probably also contained sources of sulphate (e.g. plasterboard), which may have been a major contributor to the leachate. To test this, the concentration of calcium (Ca) in the leachate was determined in order to assess the potential saturation of gypsum. A Ca concentration of  $2.83 \pm 0.36$  mmol ( $n=4$ ) with a corresponding sulphate concentration of  $6.35 \pm 2.44$  mmol ( $n=4$ ) was found in the leachate during 2003, indicating that around 45% of the sulphate in the leachate could be due to dissolution of gypsum. If we assume that the only source of Ca in the leachate was gypsum from construction waste, and that the remaining  $\text{SO}_4^{2-}$  originated from the bottom ash, this would suggest that 18% of the  $\text{SO}_4^{2-}$  deposited as ash was leached in 2002–2003.

Similar rates of mobilization of  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  from MSWI bottom ash have been described by Belevi et al. [19]. The lower discharge of  $\text{SO}_4^{2-}$  compared to  $\text{Cl}^-$  might be due to reduction to sulphide and subsequent precipitation with metals, or due to the generation of  $\text{SO}_4^{2-}$  compounds with low solubility in basic conditions [13].

In this investigation, both chloride and sulphate were measured in the initial bottom ash through water extraction. It is possible that a considerable portion of sulphates were not water soluble, and was therefore not included in the mass-balance. It is, however, the water-soluble fraction of the bottom ash that influences the leachate to the largest degree. As shown, deposited plasterboard could influence the level of sulphate considerably. The chloride level in the leachate might also be influenced by other sources at the landfill. It seems likely, however, that the deposited bottom ash represents a significant source of sulphate and chloride to the leachate. Assuming that the current water balance and permeability in the landfill are upheld, a rapid decline in the concentrations, particularly of chloride, can be expected when the deposition of MSWI bottom ash is discontinued. Such a trend was observed by Stegemann et al. [1] in a laboratory leaching experiment of bottom ash, and has also been described by Wiles [3] and Belevi et al. [19]. The decline would be less pronounced for sulphate (of which 18% was discharged per year). However, a decline in the discharged level of sulphate would be likely as the addition of easily soluble sulphates is discontinued.

#### 4. Conclusion

The mass-balance calculation in this study is an estimation of the actual mass-balance of a landfill in which the major labile component is bottom ash. The data give indication of the leaching that can be expected from landfilled MSWI bottom ash.

The retention of heavy metals was considerable, with only 0.001% (Pb) to 0.55% (Cr) of the heavy metals deposited through the bottom ash being discharged from the landfill over the same period. The retention might be overestimated compared to a bottom ash monofill, however, due to the deposition of organic material and silicates in addition to the bottom ash. On the other hand, it might be underestimated since neither the metal content for all the deposited waste, nor the metals deposited previous to 2002 had been accounted for.

Of the anions, chloride was leached readily from the landfill (80% of that added in bottom ash being leached during the same period), whereas sulphate was leached more slowly (18%), possibly due to retention as low-soluble sulphur compounds. The lack of retention of chloride in the landfill indicates that a rapid decline in the level of this anion can be expected after landfilling of MSWI bottom ash ends.

Co-disposal MSWI bottom ash and waste containing organic material does not seem to lead to higher leaching rate of heavy metals than at MSW landfills. More work is needed

to determine the potential long-term impacts of MSWI bottom ash deposition at a landfill for mixed construction waste or at a MSW landfill, however. The predicted chloride and sulphate leaching patterns should be confirmed by investigating the changes in concentrations of  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  in leachate from bottom ash monofills before, during and after closedown. The rate of leaching of heavy metals at a monofill should also be validated.

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