

Contents lists available at [ScienceDirect](http://ScienceDirect.com)

# Waste Management

journal homepage: [www.elsevier.com/locate/wasman](http://www.elsevier.com/locate/wasman)

## Development of drainage water quality from a landfill cover built with secondary construction materials

Igor Travar<sup>a,b,\*</sup>, Lale Andreas<sup>a</sup>, Jurate Kumpiene<sup>a</sup>, Anders Lagerkvist<sup>a</sup><sup>a</sup> Division of Waste Science and Technology, Luleå University of Technology, SE-971 87 Luleå, Sweden<sup>b</sup> Ragn Sells AB, Box 952, 19129 Sollentuna, Sweden

### ARTICLE INFO

#### Article history:

Received 21 March 2014

Accepted 17 September 2014

Available online 8 October 2014

#### Keywords:

Landfill cover

Ash

Sewage sludge

Drainage water

Leaching

### ABSTRACT

The aim of this study was to evaluate the drainage water quality from a landfill cover built with secondary construction materials (SCM), fly ash (FA), bottom ash (BA) sewage sludge, compost and its changes over time. Column tests, physical simulation models and a full scale field test were conducted. While the laboratory tests showed a clear trend for all studied constituents towards reduced concentrations over time, the concentrations in the field fluctuated considerably. The primary contaminants in the drainage water were  $\text{Cl}^-$ , N, dissolved organic matter and Cd, Cu, Ni, Zn with initial concentrations one to three orders of magnitude above the discharge values to the local recipient. Using a sludge/FA mixture in the protection layer resulted in less contaminated drainage water compared to a sludge/BA mixture. If the leaching conditions in the landfill cover change from reduced to oxidized, the release of trace elements from ashes is expected to last about one decade longer while the release of N and organic matter from the sludge can be shortened with about two–three decades. The observed concentration levels and their expected development over time require drainage water treatment for at least three to four decades before the water can be discharged directly to the recipient.

© 2014 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/3.0/>).

### 1. Introduction

A landfill cover is a multilayer construction that serves to reduce infiltration of water to the deposited waste and to minimize emissions of landfill gas into the atmosphere. According to Swedish legislation, a top cover must be installed after landfill closure for both municipal solid waste as well as hazardous waste landfills (SFS, 2001). A landfill cover is usually built using a combination of natural materials (e.g., gravel, sand, till and clay) and synthetic materials (e.g., geomembranes). In order to preserve natural resources and to reduce costs, re-using wastes like ashes or treated sewage sludge can be an option. However, such materials may leach contaminants as trace metals, nitrogen and organic matter.

Looking at possible pollution pathways from such a cover, two types of water can be distinguished: leachate that percolates through the whole cover construction into the waste below, and drainage water that only seeps through the layers above the liner and is collected in the drainage layer. The latter is usually discharged via a ditch at the bottom of the landfill slope. At covered landfills, apart from the surface run-off, the drainage water will

be the dominating water emission while leachate amounts are expected to be rather low. Previous studies have shown that the quality of the drainage water depends primarily on the materials installed above the liner (Travar et al., 2009). In cases where SCM are used, it is most likely that the drainage water cannot be discharged directly to the local recipient but needs at least some kind of treatment. Hence, it is important to determine the quality of the drainage water and its expected changes over time.

To be able to evaluate leaching trends, the conditions prevailing in a landfill cover must be known. Material properties, water infiltration and transport, redox potential, pH and temperature are relevant factors that affect the chemical conditions and thus the drainage water quality (Kylefors et al., 2003; Sabbas et al., 2003; van der Sloot, 1996). Changes of these factors result in changes of the chemical conditions by e.g., complex formation, acid-base reactions, redox processes, precipitation and dissolution reactions, adsorption and biochemical processes.

The main aim of this study was to evaluate the quality of drainage water generated in a landfill cover built with compost, ashes and sewage sludge as well as the development of the water quality over time. The evaluation was based on a full-scale field test, laboratory physical models (hereafter referred to as simulators) and column leaching tests. Differences between the field and laboratory tests and the influence of single layers on the drainage water

\* Corresponding author at: Division of Waste Science and Technology, Luleå University of Technology, SE-971 87 Luleå, Sweden. Tel.: +46 70 927 24 97.

E-mail address: [travar76@yahoo.com](mailto:travar76@yahoo.com) (I. Travar).

quality were discussed. Treatment needs for the drainage water were assessed based on predictions of the laboratory results in combination with the results from the field. Geochemical equilibrium calculations were used to identify mineral phases that may control the release of elements from the cover.

## 2. Material and methods

### 2.1. Full scale test

A four hectares landfill cover composed of recycled materials was established at the Tveta landfill, 30 km southwest of Stockholm, Sweden. The area was divided into six parts each using different materials. Out of these, two areas were evaluated in this paper; A1 and A4 completed in September 2003 and March 2005, respectively (Fig. 1). The areas were hydraulically separated from each other.

The foundation of the landfill cover consists of bottom ash (BA) in A1 and sand from fluid bed incinerator (FBI) in A4. This layer is connected to the gas collection system in order to avoid the accumulation of landfill gas underneath the liner. The liner consists of fly ash (FA) in A1 and a mixture of FA and BA in A4. It serves as a barrier against infiltrating precipitation water into the deposited waste and reduces landfill gas emissions. A coarse fraction of BA (>10 mm) was used in the drainage layer above the liner to collect and discharge water that percolates through the layers above. The protection layer consists of a mixture of digested and composted sewage sludge and FA in A1 and the fine fraction of BA (<10 mm) in A4. The function of this layer is to protect the liner from desiccation, freezing and root penetration. A layer of compost was placed on top of the ash-sludge mixtures in order to avoid erosion and to promote the establishment of vegetation.

The run-off from the drainage layer was sampled from wells installed in covered ditches at the bottom of the slope (Fig. 2). These wells collected drainage water from an area of about 2300 m<sup>2</sup> in A1 and 1900 m<sup>2</sup> in A4. Sampling of drainage water was performed four times per year. Totally, 20 and 9 samples of drainage water were taken from A1 and A4, respectively.

Samples of the pore water from the bottom of the vegetation layer were taken at one occasion during autumn 2008. Rhizon pore water samplers (polymeric, 10 cm long, Ø4.5 mm, medium pore size 0.1 µm, MacroRhizon, Eijkelkamp, Netherlands) were used to take four samples in A1 and A4 each. All samples were stored frozen at -20 °C prior to analysis.

The temperature in the protection layer was measured with three vertical probes installed in each area. Each probe had sensors at three depths: at the bottom, in the middle and at the top of the protection layer.

A 4		A 1		
Compost				Vegetation layer ≈ 0.3 m
Sludge/Fine BA fraction (40/60)		Sludge/FA (40/60)		Protection layer ≥ 1.5 m
Coarse BA fraction				
FA/BA (50/50)		FA		Drainage layer ≈ 0.3 m
FBI Sand		BA		Liner ≥ 1 m
				Gas drainage layer ≈ 0.3 m

Fig. 1. Design of the sub-areas A1 and A4 of the landfill cover test area at the Tveta landfill. BA = Bottom ash; FA = Fly ash; FBI = Fluid bed incineration.

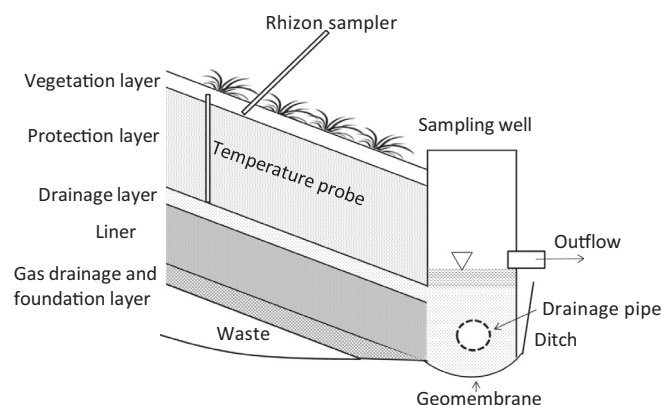


Fig. 2. Cross section of the landfill cover test area with measurement equipment.

The liquid to solid ( $L/S$ ) ratio was used for comparison of the results from the full scale test with those of the laboratory tests. The following equation was used to calculate the  $L/S$  ratio in  $l\ kg^{-1}$

$$L/S = (L/S)_0 + (I/(\rho \cdot h)) \quad (1)$$

where  $(L/S)_0$  is the initial  $L/S$  ratio,  $I$  is the net water infiltration through the material in  $l\ m^{-2}$ ,  $\rho$  is the compacted dry density of the material in  $kg\ m^{-3}$  and  $h$  is the total height of the protection and vegetation layer in m. The initial  $L/S$  ratio was calculated as the ratio between the total amount of water and the total amount of solid material in the layers above the liner. The initial  $L/S$  ratio in both areas was  $0.9\ l\ kg^{-1}$ . The  $L/S$  in the full scale test was calculated based on the actual precipitation and for a Swedish conditions typical hydrological regime assuming that about one third of the precipitation infiltrates into the ground while the rest evaporates and runs off as surface water (Raab and Vedin, 1995). A  $L/S$  of about  $0.171\ (kg\ yr)^{-1}$  was estimated which gives a cumulative  $L/S$   $1.9\ l\ kg^{-1}$  after 6 years for A1 and  $1.8\ l\ kg^{-1}$  after 5 years for A4.

### 2.2. Materials

The ashes used in the liner originate from an incineration plant with three furnaces; grate type, fluid bed and pulverized fuel incinerators. The grate type incinerator is supplied mainly with paper, plastic, wood and wood chips. The fluid bed and the pulverized fuel incinerators are fed mainly with wood and peat.

The FA is a mixture of electrostatic precipitator ash and bag house filter ash, containing among other, reacted and unreacted lime from the semi-dry flue gas cleaning process and ammonia. It was moistened prior to transport and used immediately after arrival at the landfill. When this was not possible, FA was stored covered by tarpaulins. At the time A1 was built, the FA and BA were not yet separated at the incinerator. The mixed ash was sieved at the construction site (<10 mm) which means that the used "FA" actually was the fine fraction of a FA-BA mixture containing FA predominantly. At the time A4 was built, FA and BA were separated at the incinerator and mixed in 50:50 ratio by weight at the landfill prior to construction. Most of the BA came from the grate type incinerator.

For the drainage and protection layer, BA from a municipal solid waste incineration (MSWI) plant was used. It was quenched and then stored for a couple of months for drying and aging. Afterwards, magnetic components were removed, and it was sieved through 8 mm mesh sieve. The coarse fraction was used in the drainage layer while the fine fraction was mixed with sewage sludge to build the protection layer.

The sewage sludge derived from a municipal wastewater treatment plant where it was an aerobically digested and dewatered. It

was stored for several weeks up to six months at the landfill before it was mixed with the fine fraction of the MSWI-BA.

The compost was obtained from the composting of food waste and garden and park waste.

All materials were characterised before and after the construction work (Table 1). In 2007, four and two years after the construction of A1 and A4, respectively, part of each area was excavated and samples were taken from different depths and layers. The following samples were evaluated in this study:

Prior to construction – two samples of FA and one sample of FA/BA (5–10 kg per sample), one sample of sludge/BA (10–20 kg) and one sample of drainage layer BA (10–20 kg). After construction: five samples per area from the protection layer (2–5 kg every 30 cm); two samples from the middle of the drainage layer (6–8 kg) and, two samples per area from the liner surface (1–2 kg at 2–5 cm using a drilling sampler). In 2008, samples from the middle of the vegetation layer of A1 and A4 were dug up (5–10 kg at 15 cm using a shovel) and analysed in triplicates.

### 2.3. Laboratory tests

#### 2.3.1. Simulators

Two cylindrical stainless steel containers with a volume of about 100 l each (Ø39 cm, height 84 cm, Fig. 3) were used to simulate the development of the quality of the drainage water. Chemical, biological and physical processes can be accelerated by providing favourable conditions for micro organisms, such as constant temperatures (20 °C) and homogeneous moisture distribution (Brinkmann et al., 1999).

Washed gravel was placed at the bottom of the simulators to a height of about 30 cm. A geotextile was placed over the gravel and

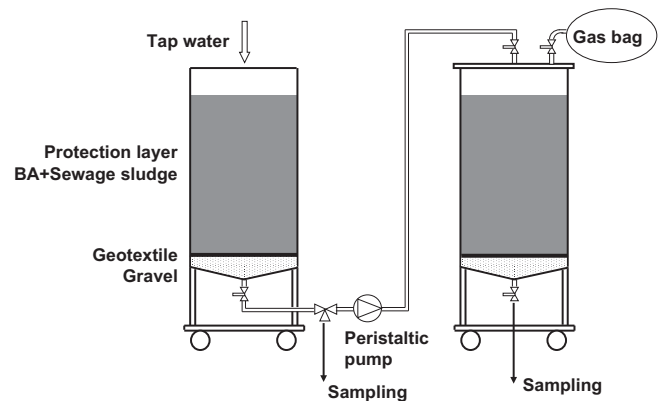


Fig. 3. Design of the physical models (simulators) used for simulation of the emissions from the protection layer material.

then both simulators were filled with the protection layer material from A4 sampled during the construction works. About 70 l of sludge/BA-P was used in each simulator, totally 160 kg (content of total solid, TS 52%). Water was added to achieve the field capacity of the material (TS 58%).

The simulators were connected in a series. The first simulator (S1) was open to the atmosphere simulating the upper part of the protection layer. The second simulator (S2) was hermetically closed simulating the lower part of the protection layer. A gas bag was attached in order to collect gas that could be formed during anaerobic digestion of residual organic matter in the sludge. About every three weeks, 5 l of tap water were added to the first simulator. After 48 h, the water that had percolated through the

**Table 1**  
Total element composition of ashes, sludge and compost used in landfill cover (P – protection layer, D – drainage layer, L – liner). Average ( $n = 3-6$ )  $\pm$  standard deviation.

	Compost A1 <sup>a</sup>	Compost A4 <sup>a</sup>	BA – P <sup>b</sup>	Sludge/FA-P A1 <sup>c</sup>	Sludge/BA-P A4 <sup>d</sup>	BA – D A1/A4 <sup>e</sup>	FA – L A1 <sup>f</sup>	FA/BA – L A4 <sup>g</sup>
<i>Major constituents g (kg TS)<sup>-1</sup></i>								
Si	n.a	n.a	140	148 $\pm$ 65	158 $\pm$ 23	227 $\pm$ 14	115 $\pm$ 19	120 $\pm$ 9.5
Al	n.a	n.a	33	21 $\pm$ 2.3	28 $\pm$ 0.8	27 $\pm$ 1.3	15 $\pm$ 6.3	20 $\pm$ 3.1
Ca	n.a	n.a	120	122 $\pm$ 44	104 $\pm$ 17	83 $\pm$ 4.7	230 $\pm$ 50	211 $\pm$ 7.3
Cl	n.a	n.a	n.a	12 $\pm$ 7	4.4 $\pm$ 1.8	7.2 $\pm$ 5 <sup>*</sup>	66 $\pm$ 10	n.a
Fe	n.a	n.a	43	28 $\pm$ 8.2	37 $\pm$ 5.1	47 $\pm$ 10	15 $\pm$ 3.7	13 $\pm$ 0.9
K	5.5 $\pm$ 0.5	7.9 $\pm$ 0.6	6.3	6.3 $\pm$ 2.6	6.1 $\pm$ 0.6	6.5 $\pm$ 0.4	4.4 $\pm$ 1.2	4.6 $\pm$ 2.3
Mg	4.7 $\pm$ 0.03	10 $\pm$ 0.5	12	11 $\pm$ 2.2	13 $\pm$ 2.1	13 $\pm$ 1.5	13 $\pm$ 3.9	11 $\pm$ 1.5
Mn	0.6 $\pm$ 0.01	1.5 $\pm$ 0.1	1.1	0.9 $\pm$ 0.1	1.6 $\pm$ 0.5	1.6 $\pm$ 0.4	1.1 $\pm$ 0.05	1.1 $\pm$ 0.1
Na	n.a	n.a	16	8.7 $\pm$ 2.2	11 $\pm$ 2.9	19 $\pm$ 2.2	3.3 $\pm$ 0.7	5.6 $\pm$ 1.4
N	n.a	n.a	n.a	5.1 $\pm$ 1.7	2.2 $\pm$ 0.4	n.a	n.a	n.a
P	7286 $\pm$ 292	9.6 $\pm$ 1.3	4.2	6.7 $\pm$ 1.5	5.4 $\pm$ 0.6	1.9 $\pm$ 0.1	1.0 $\pm$ 0.2	1.2 $\pm$ 0.1
S	3.9 $\pm$ 0.4	4.2 $\pm$ 0.2	12	14.9 $\pm$ 9.1	7.5 $\pm$ 2.7	3.6 $\pm$ 0.3	30 $\pm$ 9.2	27 $\pm$ 4.8
<i>Minor constituents mg (kg TS)<sup>-1</sup></i>								
As	21 $\pm$ 3.8	1.6 $\pm$ 0.3	31	52 $\pm$ 18	26 $\pm$ 6.7	23 $\pm$ 6.3	136 $\pm$ 19	125 $\pm$ 7.1
Ba	n.a	n.a	1290	1135 $\pm$ 166	1496 $\pm$ 638	1203 $\pm$ 146	1722 $\pm$ 274	2300 $\pm$ 191
Cd	42 $\pm$ 2.7	187 $\pm$ 20	7.6	7.2 $\pm$ 3.3	16 $\pm$ 9.3	8.3 $\pm$ 1.1	13 $\pm$ 1.1	28 $\pm$ 2.7
Cr	194 $\pm$ 70	345 $\pm$ 34	455	348 $\pm$ 55	672 $\pm$ 170	1585 $\pm$ 791	328 $\pm$ 75	480 $\pm$ 36
Cu	3545 $\pm$ 2619	3994 $\pm$ 317	3190	2109 $\pm$ 984	2535 $\pm$ 455	1913 $\pm$ 296	1171 $\pm$ 998	3197 $\pm$ 488
Hg	n.a	n.a	0.4	0.6 $\pm$ 0.2	1.0 $\pm$ 0.7	0.2 $\pm$ 0.2	1.2 $\pm$ 0.5	1.4 $\pm$ 0.02
Mo	n.a	n.a	15	11 $\pm$ 2.4	22 $\pm$ 8.9	24 $\pm$ 15	12 $\pm$ 2.5	26 $\pm$ 11
Ni	268 $\pm$ 366	304 $\pm$ 113	180	74 $\pm$ 16	195 $\pm$ 71	455 $\pm$ 194	61 $\pm$ 26	112 $\pm$ 23
Pb	368 $\pm$ 52	2133 $\pm$ 2032	883	765 $\pm$ 320	896 $\pm$ 282	993 $\pm$ 451	1450 $\pm$ 460	2257 $\pm$ 98
Zn	2058 $\pm$ 499	3949 $\pm$ 274	9480	2382 $\pm$ 514	4332 $\pm$ 1075	2410 $\pm$ 450	2976 $\pm$ 440	5807 $\pm$ 653
LOI	n.a	n.a	132	212 $\pm$ 55	166 $\pm$ 34	45 $\pm$ 7.3	164 $\pm$ 18	167 $\pm$ 17

n.a. data not available.

LOI – loss on ignition.

<sup>a</sup> 3 excavated samples.

<sup>b</sup> 1 sample taken before construction works.

<sup>c</sup> 5 excavated samples.

<sup>d</sup> 5 excavated samples plus 1 sample before construction works.

<sup>e</sup> 2 excavated samples plus 1 sample before construction works.

<sup>f</sup> 2 excavated samples plus 3 samples before construction works.

<sup>g</sup> 2 excavated plus 1 sample before construction works.

<sup>\*</sup> Concentration of Cl<sup>-</sup> in the BA which originates from the same incineration plant found in Avfall Sverige (2011) report.

material in the first simulator was pumped to the second simulator. After about two weeks, both simulators were discharged and left to rest for a week. The infiltration regime was chosen to provide sufficient water for biological processes but at the same time to avoid flushing the material and to simulate periods with and without precipitations (intermittent wetting). This irrigation scheme corresponds to a rate of 2–2.5 m<sup>3</sup> per metric ton protection layer material and year, whereas in the field test the rate is about 0.17 m<sup>3</sup> (t yr)<sup>-1</sup>. Thus, the water balance in the simulation reactors was about 12–15 times higher than at full-scale landfill. The initial *L/S* ratio in both simulators was 0.9 l kg<sup>-1</sup>, and final *L/S* ratio reached at the end of this test was 5.5 l kg<sup>-1</sup>. Water samples were taken from both simulators about every *L/S* 0.15. After measuring pH, redox potential and electrical conductivity, the samples were stored at -20 °C prior to further analysis. The influence of the vegetation layer, drainage layer and liner surface on the drainage water quality was not simulated in this test.

### 2.3.2. Column test

A column leaching test was performed in triplicates according to SIS-CEN/TS 14405 (up-flow percolation test). The excavated samples taken from the middle of the protection layer from A1 and A4 were used in this test. The initial *L/S* ratios were 1.7 l kg<sup>-1</sup> in column A1 and 1.5 l kg<sup>-1</sup> in column A4. The drainage water was sampled at cumulative *L/S* ratios of 1.8, 2, 3, 4.2, 7.3 and 19.

### 2.4. Mobilisation of elements in the field and laboratory tests

The following equation was used to calculate the mobilised fraction (MF) of an element in % or in ‰ of total element concentration in the field test from the layers above the liner

$$MF = C_1 \cdot (I / (\rho \cdot h)) / C_2 \quad (2)$$

where  $C_1$  is the concentration of an element in the drainage water in mg l<sup>-1</sup>, and  $C_2$  is the total concentration of an element in the layers above the liner in mg (kg TS)<sup>-1</sup>.

The MF of an element in the laboratory tests was calculated as

$$MF = C_1 \cdot (I_1 / m_{dry}) / C_3 \quad (3)$$

where  $I_1$  is the volume of water in l added to simulators or columns,  $m_{dry}$  is the mass of dry material in kg TS, and  $C_3$  is total concentration of an element in simulators and columns expressed as mg (kg TS)<sup>-1</sup>.

### 2.5. Chemical analysis

The elemental composition of the solid sampled material was determined at an accredited laboratory using ICP-AES (EPA method 200.7) and ICP-SFMS (EPA method 200.8; both methods modified according to the instruments used and Swedish legal guidelines) after digestion according to the ASTM methods D3683 and D3682. Loss on ignition (LOI) was determined after ignition at 1000 °C for 2 h.

Aqueous samples were analysed for element concentrations using ICP-SFMS (EPA method 200.8; modified) without prior digestion. Chloride, total nitrogen ( $N_{tot}$ ) and ammonia nitrogen ( $NH_4-N$ ) were analysed spectrophotometrically (AAE Quaatro, Bran + Luebbe, Germany);  $N_{tot}$  after digestion according to Swedish standard SS 028131 (SIS, 1976). The pH, electrical conductivity, and redox potential were analysed immediately after sampling. Total organic carbon (TOC) was determined according to EN 1484 (CEN, 1997) with a TOC analyser (TOC-V CSH Shimadzu).

### 2.6. Geochemical modelling

Geochemical modelling was performed using Visual Minteq version 2.61. The objective was to: (i) identify potential solubility controlling minerals, (ii) evaluate the influence of organic matter on the leaching of elements and (iii) study the effect of CO<sub>2</sub> partial pressure on the drainage water chemistry. The minerals discussed were selected based on: (i) their likeness to be present or formed in MSWI ashes (according to e.g., Dijkstra et al., 2006; Johnson et al., 1999; Zevenbergen et al., 1996) and (ii) calculated saturation indices (SI) that approach zero ( $-1 < SI < 1$ ). The SI indicates if a solution is undersaturated ( $SI < 0$ ) or oversaturated ( $SI > 0$ ) with respect to the solid. When the SI is close to zero, there is an apparent equilibrium with respect to the solid.

The input files were composed of measured element concentrations, fixed pH to the measured value, alkalinity, temperature and redox potential of the drainage water sampled from the field and simulators. Concentrations of Fe were assumed to be as Fe<sup>2+</sup> when redox is negative while Fe<sup>3+</sup> was assumed in calculations under positive redox conditions. Complexation of trace elements with dissolved organic matter was evaluated using the Stockholm humic model (SHM). It was assumed that 70% of the active solid-phase organic matter consisted of humic acid (HA), whereas 30% was fulvic acid (FA) (Gustafsson and Berggren Kleja, 2005). Activities of species were calculated using the Davies equation (Gustafsson and Berggren Kleja, 2005). Partial pressure of CO<sub>2</sub> was varied from  $3.8 \times 10^{-4}$  atm to  $3.8 \times 10^{-2}$  atm to evaluate the possible influence of landfill gas that may diffuse through the liner on drainage water chemistry.

## 3. Results

### 3.1. Total element concentrations

Table 1 shows the total composition of the materials used in the landfill cover. The ashes were relatively rich in Zn, Cu, Pb, Cr and As. The fine fraction of the MSWI BA (BA-P) had the highest concentrations of Cu and Zn. Most trace elements in the sludge/BA-P mixture originated likely from the BA-P while the sludge contributed to the elevated content of N and organic matter in the mixture (as indicated by the LOI). The coarse fraction of the MSWI BA (BA-D) contained elevated concentrations of Cr and Ni compared to the other ashes while the ashes used in the liner showed high concentrations of As, Cu, Pb and Zn. The compost used in the vegetation layer contained a high concentration of P in A1, elevated levels of Cd and Cu in both areas and high Pb and Zn in A4 in comparison with sludge and ashes.

### 3.2. Leaching conditions

The pH values of the drainage water in the field test varied between 7 and 8.6 while the laboratory tests showed pH between 6.9 and 8.1 (Fig. 4). The difference between pH of drainage water in the field test and laboratory tests at similar *L/S* ratios was found not to be significant ( $\alpha = 0.05$ ). The pH difference between the simulators S1 and S2 was significant ( $\alpha = 0.05$ ).

The temperature in the field varied over time, following the ambient air temperature (Fig. 5). The variations were between 17 and 27 °C at the bottom (B) and 2–30 °C at the top (U) of the protection layer. The temperature in the laboratory tests was relatively constant at ~20 °C, which means that the difference between the field and laboratory tests was in the range of 5–20 °C. Despite outside temperatures close to -10 °C during winter, no freezing was observed in the protection layer (Fig. 5).

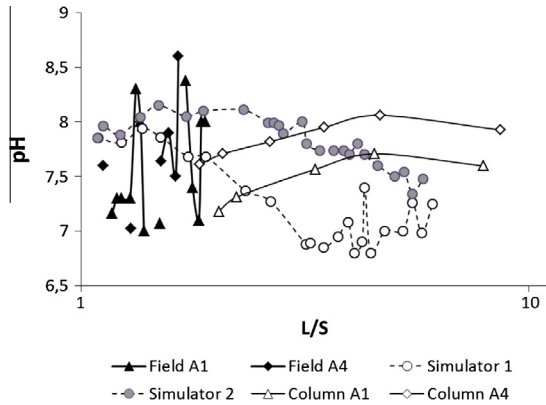


Fig. 4. Development of pH in the field (Areas A1, A4), simulators and column test (columns A1 and A4).

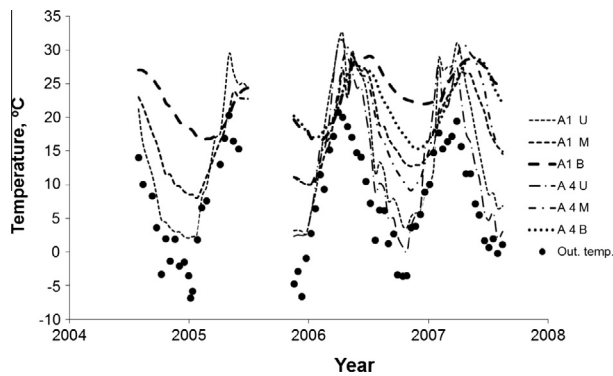


Fig. 5. Temperature in the protection layer measured at three depths from 2004 to 2008; U – upper (close to ground surface), M – middle, B – bottom; Out. temp. – outside air temperature.

### 3.3. Leaching of constituents in laboratory and field

Figs. 6 and 7 illustrate the mobilization of  $\text{Cl}^-$ , N-tot, As, Cd, Cr, Cu, Ni, Mo, Pb and Zn over time. These constituents were chosen based on the quality requirements that drainage water has to fulfil before discharged to the local recipient (see Table 2). The mobilisation of the constituents is presented as cumulative percentage or per mill of the total concentration in the layers above the liner as a function of  $L/S$  ratio. A decrease of the slope of the curves corresponds to a decrease in leaching rate (Figs. 6 and 7).

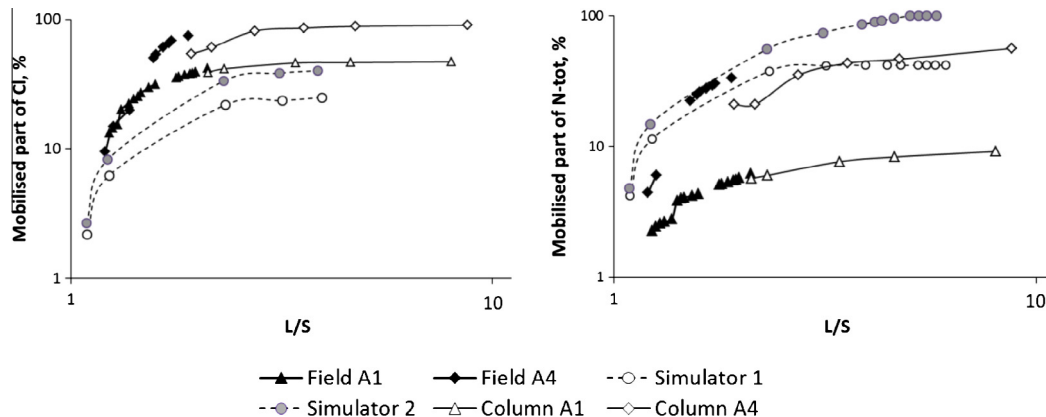


Fig. 6. Cumulative mobilised fraction (%) of the total element concentration of  $\text{Cl}^-$  (a) and  $\text{N}_{\text{tot}}$  (b) from the material in the field, simulators and column tests as a function of  $L/S$ .

Since the concentrations of  $\text{Cl}^-$  and N were not analysed in the compost and in the drainage layer material (BA-D), it was assumed that the compost contained about the same concentrations of these elements as the protection layer material. The concentration of  $\text{Cl}^-$  in BA-D was assumed to be the same as in BA measured in previous studies at the same incineration plant ( $7.2 \pm 5 \text{ mg kg}^{-1}$ , see Table 1) (Avfall Sverige, 2011) while the concentration of N was assumed as negligible in BA-D.

$\text{Cl}^-$  was washed out at an  $L/S$  ratio of about  $3 \text{ l kg}^{-1}$  in the laboratory tests (Fig. 6a). The drainage layer and liner were absent in the laboratory tests and only the protection layer without any adjacent materials was tested. The higher mobilisation of  $\text{Cl}^-$  in the field can be explained by the impact of the liner and drainage layer on the  $\text{Cl}^-$  concentrations in the drainage water. Alternatively, the total amount of  $\text{Cl}^-$  in the field was underestimated. The release of  $\text{Cl}^-$  was positively correlated with the leaching of Cd (correlation coefficients were between 0.72 in S1 and 0.93 in S2).

The leached amounts of trace elements were low compared to the total amounts in the used materials. For example, the mobility varied between 0.1‰ for Cr and Pb and about 90‰ for Mo (Fig. 7). The leaching of trace elements apart from Cd, Ni and Zn decreased during the test period in S1 and columns ( $L/S$  5.5 and 7.8 respectively). In S2, the leaching more or less ceased at the latest at  $L/S = 3.5 \text{ l kg}^{-1}$ , indicating that these elements may be present in the drainage water only for a short period.

The concentrations of TOC in the drainage water decreased in the laboratory tests (Fig. 8) and a positive correlation with the pH in the simulators could be seen (correlation coefficients were 0.93 in S1 and 0.4 in S2). A part of TOC in the drainage water from S2 originated from S1, which can explain the higher concentrations of TOC in S2 compared to S1. Lower concentrations of TOC in the field test compared to simulators were probably related to higher degradation rate of organic matter in the field.

Table 2 illustrates the changes in concentrations of constituents over  $L/S$  in the drainage water from the field, simulators and column test. The term “Limit” represents the discharge criteria for the drainage water at the Tveta landfill. The concentrations in the field exceeded the limit for all elements except for  $\text{NH}_4\text{-N}$  in both areas and As, Cr and Pb in A1 at  $L/S = 2 \text{ l kg}^{-1}$ . Results from the laboratory tests show that the discharge limits for N were reached at  $L/S = 3 \text{ l kg}^{-1}$  in S1 while it still leached in S2 after  $L/S = 5.5 \text{ l kg}^{-1}$  (Table 2 and Fig. 6b). The leaching trend of TOC in the laboratory tests showed that TOC might be present in the drainage water even after  $L/S = 5.5 \text{ l kg}^{-1}$  (Fig. 8 and Table 2). Low release of the trace elements was observed in S2 after  $L/S = 4.2 \text{ (kg yr)}^{-1}$  while Cd, Cu, Ni and Zn were still present in the drainage water of S1 in concentrations above the limit even after  $L/S = 5.5 \text{ l (kg yr)}^{-1}$ .

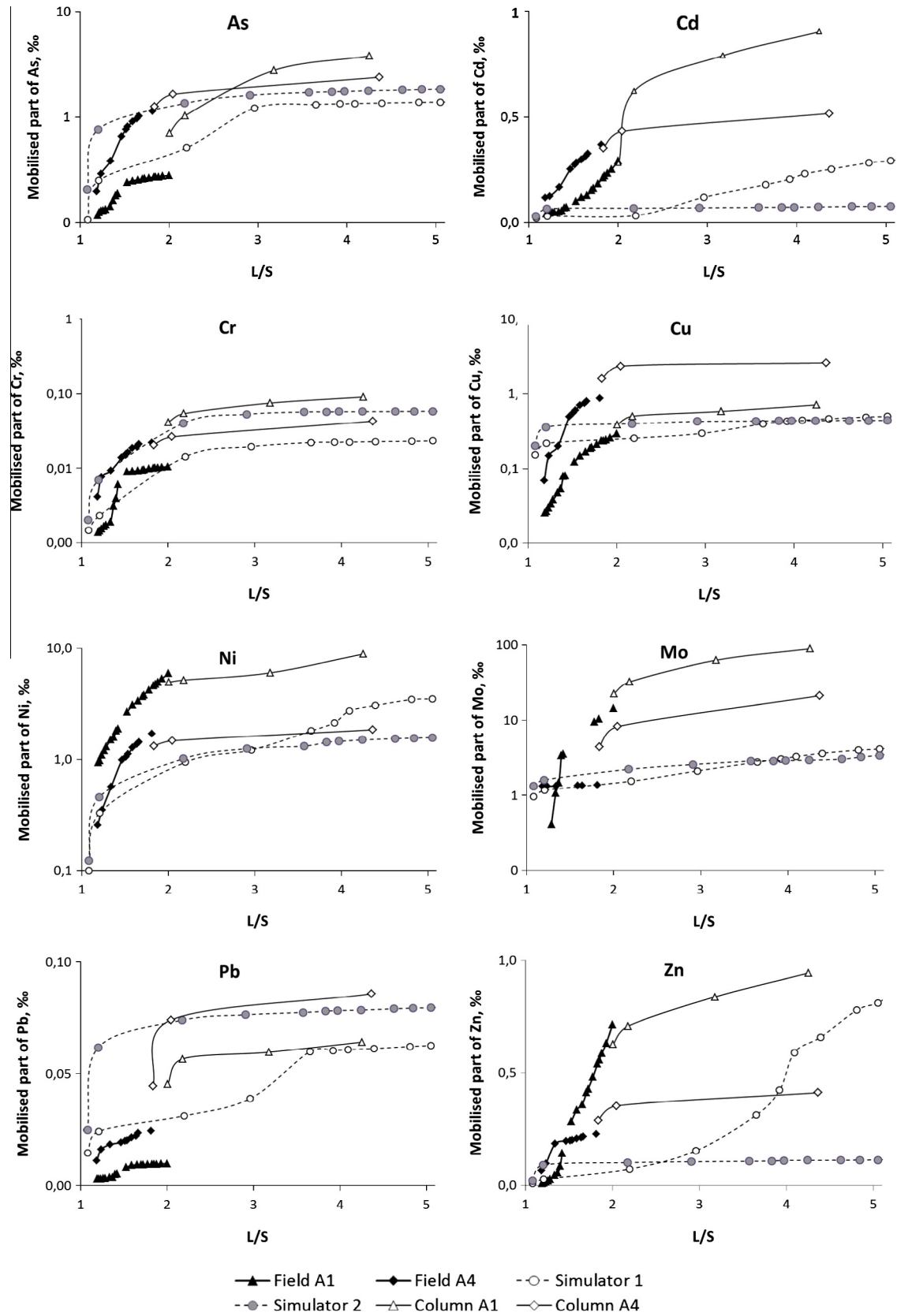


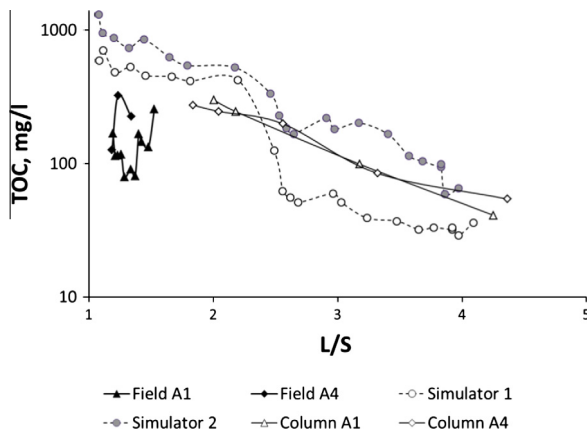
Fig. 7. Cumulative mobilised fraction (%o) of the total element concentration in the field, simulators and column tests as a function of L/S.

Nitrogen in the drainage water from the laboratory tests occurred mainly as  $\text{NH}_4\text{-N}$  while a higher portion of oxidized species were present in the drainage water from the field (Table 2).

This indicates that the redox potential was higher in the field or that the drainage water was oxidized before sampling. Both  $\text{NH}_4\text{-N}$  and  $\text{N}_{\text{tot}}$  concentrations went very low in S1 (open to the

**Table 2**  
Concentrations of constituents at different *L/S* in the drainage water from the field, simulators and column tests compared with the limit values for direct discharge to the local recipient at Tveta landfill. *Italic numbers indicate values below the discharge limits.*

Element Unit	NH <sub>4</sub> -N mg l <sup>-1</sup>	N-tot mg l <sup>-1</sup>	TOC mg l <sup>-1</sup>	As μg l <sup>-1</sup>	Cd μg l <sup>-1</sup>	Cr μg l <sup>-1</sup>	Cu mg l <sup>-1</sup>	Ni μg l <sup>-1</sup>	Pb μg l <sup>-1</sup>	Zn μg l <sup>-1</sup>
<i>L/S = 1.1</i>										
Field A1	166	413	114	20	0.8	1	0.1	190	2	339
Field A4	48	448	126	40	15	22	1.3	390	75	2100
Simulator 1	1 100	1 100	480	32	1.5	5	1.3	380	73	590
Simulator 2	1 800	1 800	870	130	5.1	30	3.4	580	290	2500
<i>L/S = 1.8</i>										
Field A4	0.7	286	224	21	5.1	5.3	1.4	379	4.8	315
<i>L/S = 2</i>										
Field A1	0.2	130	256	3.6	2.9	1	0.7	560	1	2300
Column A1	49	94	245	98	14	26	1.5	92	49	1105
Column A4	548	650	245	52	6.8	21	9.7	150	135	1350
Simulator 1	193	209	422	7.5	1.5	9	0.1	130	7	200
Simulator 2	623	629	524	17	0.05	25	0.1	120	12	51
<i>L/S = 4.2</i>										
Column A1	30	32	41	50	0.75	5	0.3	199	3.2	235
Column A4	109	64	54	9.1	0.6	5.2	0.3	34	4.8	110
Simulator 1	0.01	1.3	37	1.4	1.4	0.9	0.2	227	1.5	997
Simulator 2	231	325	60	2.99	0.05	0.5	0.01	35	0.7	6.6
<i>L/S = 5.5</i>										
Simulator 1	0.005	1	n.a.	1.2	0.9	1	0.1	96	2	738
Simulator 2	92	119	n.a.	2	0.05	1	0.005	14	1.2	15
Limit	5	10	12	10	0.2	15	0.009	30	2	30



**Fig. 8.** Concentrations of TOC in drainage water over time in the field, simulators and column tests.

atmosphere), mainly due to the wash-out but also nitrification process. The higher concentrations of nitrogen species in S2 even after *L/S* 5.5 were probably caused by the supply with the water from S1 but also by limited nitrification/denitrification processes as S2 was closed towards the atmosphere.

#### 3.4. The pore water quality in the vegetation layer

Table 3 shows the chemical composition of the pore water from the vegetation layer in the field test and from vegetation layers built with conventional materials such as excavated soil (moraine and clay-moraine) or a mixture of excavated soil and sewage sludge found in the literature (Kumpiene and Lagerkvist, 2009). The concentrations of elements in the pore water were within the concentration intervals given in the literature (Kumpiene and Lagerkvist, 2009).

Compared with drainage water sampled at the same time when the pore water was sampled (corresponding to *L/S* = 2 l kg<sup>-1</sup>), the element concentrations were significantly ( $\alpha < 0.05$ ) lower in the

**Table 3**  
The chemical composition of the pore water collected in the vegetation layer.  $n = 3$ ,  $\pm$  standard deviation.

Unit	Area		Literature
	A1	A4	Kumpiene and Lagerkvist (2009)
pH	8 ± 0.1	8 ± 0.1	4.6–8.3
EC mS cm <sup>-1</sup>	0.9 ± 0.4	4.7 ± 0.7	0.04–7.1
TOC mg l <sup>-1</sup>	50 ± 20	60 ± 8	
Al μg l <sup>-1</sup>	19 ± 13	0.9 ± 0.2	<0.3–13,000
As μg l <sup>-1</sup>	<9 ± 0	<9 ± 0	0.9–13
Cd μg l <sup>-1</sup>	<0.7 ± 0	<0.1 ± 0	<0.1–24
Cr μg l <sup>-1</sup>	1.3 ± 0.5	0.1 ± 0	<0.1–3
Cu μg l <sup>-1</sup>	248 ± 72	219 ± 92	<0.5–1000
Fe μg l <sup>-1</sup>	15 ± 11	10 ± 13	<0.1–42,000
Mn μg l <sup>-1</sup>	1.1 ± 0.7	20 ± 16	0.2–8600
Ni μg l <sup>-1</sup>	38 ± 22	58 ± 22	<0.1–340
Pb μg l <sup>-1</sup>	5.8 ± 2	6.5 ± 8.3	<3–25
Zn μg l <sup>-1</sup>	229 ± 214	148 ± 53	2–9200

pore water, except for Pb in A4 (Table 2 and 3). In the simulators, Cu and Zn were lower compared to the pore water. It indicates that the vegetation layer contributed to the content of Pb, Cu and Zn to the drainage water quality.

#### 3.5. Geochemical modelling

The calculated SI indicated that calcite (CaCO<sub>3</sub>) (for all samples except for A1), smithsonite (ZnCO<sub>3</sub>) and NiCO<sub>3</sub> (except for S2 after *L/S* of 3 l kg<sup>-1</sup>), gibbsite (Al(OH)<sub>3</sub>) (for S2), aloxite (Al<sub>2</sub>O<sub>3</sub>) (in the field and S1), cerussite (PbCO<sub>3</sub>) and Pb(OH)<sub>2</sub> (in the simulators at *L/S* of 1 l kg<sup>-1</sup>) could be in equilibrium with the drainage water (Table 4). The simulations indicated that several Fe/Al-(hydr) oxides (e.g., Al(OH)<sub>3</sub>, gibbsite; AlOOH, diaspore; Fe(OH)<sub>3</sub>, ferrihydrite; FeOOH, goethite; Fe<sub>2</sub>O<sub>3</sub>, hematite) and amorphous aluminosilicates (e.g., Al<sub>2</sub>SiO<sub>3</sub>(OH)<sub>4</sub>, imogolite; Al<sub>2</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub>, kaolinite; Al<sub>2</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub> × 2H<sub>2</sub>O, halloysite) could precipitate. The drainage water was mostly undersaturated with respect to hydroxides of trace elements except for Pb(OH)<sub>2</sub> in the simulators at *L/S* = 1 l kg<sup>-1</sup>.

**Table 4**Saturation indices (SI) for minerals in drainage water from the landfill cover test areas A1 and A4 and simulator test. Italic numbers indicate SI that approach zero ( $-1 < \log SI < 1$ ).

<i>L/S, l kg<sup>-1</sup></i>		<i>log SI</i>							
Element	Mineral	Field A1	Field A4	Simulator 1			Simulator 2		
		1.1	1.1	1	3	5	1	3	5
<i>Major elements</i>									
Al	Al(OH) <sub>3</sub> , Gibbsite	1.1	1.8	2.2	2	0.9	2.5	0.8	0.8
	AlOOH, Diaspore	0.8	1.5	1.9	1.7	0.6	2.2	0.5	0.5
	Al <sub>2</sub> O <sub>3</sub> , Aloxite	-0.9	0.5	1.3	0.9	-1.2	2	-1.4	-1.5
	Al <sub>2</sub> SiO <sub>3</sub> (OH) <sub>4</sub> , Imogolite	n.a.	n.a.	4.2	4.2	2	5.1	1.8	1.9
	Al <sub>2</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub> , Kaolinite	n.a.	n.a.	6.1	6.5	4.1	7.1	3.9	4.1
	Al <sub>2</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub> × 2H <sub>2</sub> O, Halloysite	n.a.	n.a.	4	4.3	2	5	1.7	2
Ca	CaCO <sub>3</sub> , Aragonite	2.6	0.7	-0.2	0.3	0.2	0.4	0.2	0.4
	CaCO <sub>3</sub> , Calcite	2.7	0.9	-0.06	0.4	0.4	0.6	0.4	0.5
Fe	FeOH <sub>2</sub> , cristalline	8	7.8	9.3	7.3	n.a.	9.2	7.3	n.a.
	Fe(OH) <sub>3</sub> , Ferrihidrite	4.5	4.1	5.8	3.8	2.4	5.6	4	4.5
	FeOOH, Goethite	7.2	6.8	8.5	6.5	5.1	8.3	6.7	7.2
	Fe <sub>2</sub> O <sub>3</sub> , Hematite	17	16	19	15	12	19	15	17
<i>Trace elements</i>									
Cd	Cd(OH) <sub>2</sub>	-6.8	-5.1	-6.4	-7.9	-8	-6.3	-8.3	-8.2
	CdCO <sub>3</sub> , Otavite	-1.2	-2.3	-2	-1.8	-2.2	-1.6	-3.2	-2.9
Cr	Cr(OH) <sub>3</sub>	-4.2	-4.3	-4.4	-6	-6.8	-4.2	-5.1	-6.3
Cu	Cuprite, Cu <sub>2</sub> O	-17	-16	-19	2.5	3.6	n.a.	-21	n.a.
	CuCO <sub>3</sub>	0.04	-1.1	-1.3	-1.6	-2.5	-0.9	-1.4	-1.6
Mo	PbMoO <sub>4</sub> , wulfenite	-2.7		1.3	-0.2	-1.5	1.5	-3.1	-2.8
	CaMoO <sub>4</sub> , Powellite	-1.3	-0.7	-1	-2	-1.7	-0.6	-2.7	-2.7
Ni	NiOH <sub>2</sub> (cristalline)	-1.1	-1.8	-3.5	-4.8	-2.7	-1.4	-4.6	-5
	NiCO <sub>3</sub>	0.9	-0.1	-0.7	-0.3	-0.6	-0.4	-1.1	-1.2
Pb	Pb(OH) <sub>2</sub>	-3.3	-2.4	-0.8	-2.5	-3.9	-0.7	-3.8	-3.6
	PbCO <sub>3</sub> , Cerussite	-1.9	-1.3	-0.8	-0.8	-2.4	-0.3	-2.9	-2.5
Zn	Zincite, ZnO	n.a.	-1.6	-1.3	-2.3	-2.4	-1.1	-3.3	-3.8
	Zn(OH) <sub>2</sub> am	-2.6	-2.8	-2.6	-3.5	-3.6	-2.4	-4.5	-5.1
	ZnCO <sub>3</sub> , Smithsonite	0.7	0.3	-0.6	0.1	-0.1	-0.05	-1.8	-2.6

n.a. data not available.

The major result from varying the CO<sub>2</sub> partial pressure was that the SI of carbonates, e.g., calcite (CaCO<sub>3</sub>), dolomite (CaMg(CO<sub>3</sub>)<sub>2</sub>), huntite (CaMg<sub>3</sub>(CO<sub>3</sub>)<sub>4</sub>), otavite (CdCO<sub>3</sub>), siderite (FeCO<sub>3</sub>), nickel(II)carbonate (NiCO<sub>3</sub>), cerussite (PbCO<sub>3</sub>) and smithsonite (ZnCO<sub>3</sub>) increased when the partial pressure of CO<sub>2</sub> was increased from  $3.8 \times 10^{-4}$  atm to  $3.8 \times 10^{-2}$  atm. It means that the increase of the partial pressure of CO<sub>2</sub> enhanced the formation of these carbonates. No effects on Al hydr(oxides) could be seen.

## 4. Discussion

### 4.1. Influence of landfill cover components on the drainage water quality

The vegetation layer seemed to have influence on drainage water composition through the release of Cu, Pb and Zn (Table 2 and 3). Plants may reduce the mobility of these elements in the vegetation layer through sorption by roots, precipitation and complexation. Alternatively, the release of these elements can be increased when plant roots secrete substances which acidify the rhizosphere and decrease pH of the soil (Ali et al., 2013; Tao et al., 2004; Yoon et al., 2006). Moreover, root exudates may affect the release of Cu, Pb and Zn: directly by affecting the redox reactions or indirectly through their effects on microbial activity and root growth (Kidd et al., 2009; Tao et al., 2004). The release of trace elements from compost was low compared to conventional materials (Table 3) which indicated that compost can be used beneficially in the vegetation layer.

The protection layer with about 1.5 m thickness and its chemical composition obviously have the largest impact on the quality of the drainage water (Table 1). The lower concentrations observed

for almost all trace elements and TOC in the drainage water of A1 as compared to that of A4 in the field test were probably a result of using FA-P instead of BA-P as a component in the ash/sludge mixture (Table 2 and Fig. 8). The FA-P had a larger specific surface area than the BA-P, which provided more surfaces where the contaminants were adsorbed (Karlfeldt and Steenari, 2007). Moreover, a large part of the unburned carbon in the FA-P can be expected to hold properties similar to those of active carbon which gives a high adsorption capacity for both inorganic and organic substances (Bjurström et al., 2014). Hence, the use of a sludge/FA-P mixture instead of a sludge/BA-P mixture might be a better option for the protection layer.

During run-off, the drainage water is in contact with the liner surface. When water runs over a compacted surface, molecular diffusion and surface dissolution mechanisms are the dominant processes determining element release from the liner (Kosson et al., 1996; Sabbas et al., 2003). The differences in element release between the experiments indicate that the liner surface may contribute with Cl<sup>-</sup>, Cd, Ni and Zn to the drainage water (see Figs. 6 and 7). Easily soluble species like Cl<sup>-</sup> will leach from the liner surface to L/S of 2–3 l kg<sup>-1</sup> (Fig. 6a). A release of metal cations from the liner might also occur in the long term as long as concentration gradients exist in the liner. Diffusion of elements through the liner orientated towards the surface is in the opposite direction to the hydraulic gradient. When drainage water percolates through the liner, the dominant transport of the trace elements is orientated towards the landfill interior. The liner might contribute to the quality of drainage water via diffusion only when the percolation rate is low e.g., during drier periods. Thus, the drainage water is probably more contaminated due to release of contaminants from the liner after dry periods.



#### 4.2. Development of the drainage water quality over time

Ashes and sludge in a landfill cover are exposed to chemical weathering that influences the leaching of elements and hence the quality of the drainage water over time. The release of contaminants from ashes and sludge as a function of the *L/S* ratio was used to assess changes in the drainage water quality over time.

The pH fluctuated in the field test and no trends could be seen yet (Fig. 4). Fluctuations of the pH in the field could be explained by the influence of the drainage layer and liner surface on the drainage water during wet/dry cycles or due to differently long contact time before sampling between landfill gas or air and drainage water in the sampling well. Lower values of pH in S1 compared to S2 (Fig. 4) were probably due to uptake of CO<sub>2</sub> from the atmosphere and carbonation of BA and due to oxidation of the nitrogen (Table 2). The near neutral pH in the drainage water from S1 after *L/S* = 2 indicated that an equilibrium between the bigger pores of ash/sludge mixture and atmospheric CO<sub>2</sub> was reached. The change of the drainage water pH can be controlled by Ca minerals such as calcite but soluble Al and sulphates also play a role (Johnson et al., 1995; Meima and Comans, 1998). According to the equilibrium calculations (Table 4), calcite was close to equilibrium in the drainage water from A4 and in the simulators which mean that calcite could control the pH in these tests. In the field, the diffusion of landfill gas through the liner will result in the absorption of CO<sub>2</sub> by the ash leading to a slow decrease of the pH towards neutral conditions as it was observed in simulators. The pH of the drainage water will most likely stay close to neutral over extended time, due to carbonation of the ashes and biological degradation of sewage sludge.

Carbonation of the BA and decrease of pH to neutral values in drainage water (Fig. 4) will change the mineralogical characteristics of the BA matrix and hence leaching of metal cations such as Pb, Zn and Ni from BA (Dijkstra et al., 2006). Mineralogical changes of BA include dissolution and formation of mineral phases that may control the release of elements or lead to the formation of reactive sorptive solid substances which can bind trace elements. According to saturation indices, carbonates of Zn, Pb and Ni might control the release of these elements from BA/sludge in the field, in S1 up to *L/S* 2.6 and in S2 at the beginning of the experiment (Table 4). The formation of carbonates had a positive impact on the drainage water quality since these minerals are less soluble and more stable than oxides and hydroxides of these trace elements (Brännvall, 2013). Moreover, the carbonation of BA results in an agglomeration of ash particles and clogging of the ash pores which reduces the specific surface area for leaching and decreases the hydraulic conductivity of the material (Chandler et al., 1997; Brännvall, 2013; Maurice and Lagerkvist, 1998). A reduced hydraulic conductivity of the protection layer may result in more surface water on top of the landfill cover and less drainage water. Carbonation might also cause clogging of the drainage layer and increase of hydraulic head above the liner which might result in increased amounts of leachate.

The vegetation layer and also to some extent the upper parts of the protection layer are exposed to the atmosphere and thus, aerobic conditions are likely to prevail. Landfill gas that diffuses through the liner can create anaerobic conditions in the drainage layer and in the lower part of the protection layer. The change from oxidized to reduced environment in the simulator test resulted in the lower release of Ni, Cd and Zn and in the higher leaching of As (Table 2 and Fig. 7). Low redox potential in the lower parts of the protection layer may result in longer periods of high N concentrations in the drainage water (Table 2). Oxidized conditions in the protection layer will prevent dissolution of Fe oxides which keep As immobilised (Kumpiene et al., 2009) and it will reduce N concentrations in the drainage water. In the case of Ni, Cd and Zn,

reducing atmosphere in the protection layer is preferable to decrease leaching of these elements (Kosson et al., 1996; van der Sloot, 1996). If the liner gets damaged, landfill gas could be released to the upper layers of the landfill cover creating an anaerobic environment or in another case, collection system for the landfill gas might suck the air to the landfill causing aerobic conditions in a landfill cover. In the first case, it could amplify the immobilization of cationic elements, but might cause mobilisation of oxyanions such as As and in the second case vice versa. Furthermore, development of vegetation followed by associated microorganisms might lead to accumulation of organic matter in the vegetation layer and hence a decrease in the redox potential. While a lower rate of landfill gas production, and hence a lower transport of landfill gas through the liner, might counteract the redox decrease over time. The material porosity and the thickness of the layers are also important factors controlling the redox development in landfill covers (Kumpiene et al., 2013).

After the summer, the release of Cl<sup>-</sup> increased about 30–50% due to the higher temperature in the upper part of the protection layer (Fig. 5). Petrucci (1985) showed that the solubility of salts increases about 10% when the temperature rises from 20 to 30 °C. It is expected that the temperature in the protection layer will decrease over time due to the lower rate of waste decomposition, less landfill gas production and thus less methane oxidation in the protection layer. This will cause the solubility of Cl<sup>-</sup> and trace elements to reduce over time (Kylefors et al., 2003).

The release of Cl<sup>-</sup> was very intensive in the field and in the simulators (Fig. 6), which shows that the leaching of Cl<sup>-</sup> from ashes was availability controlled. The strong positive correlation between leached Cl<sup>-</sup> and Cd indicates that Cd was probably mobilised by Cl<sup>-</sup>. As described by van der Sloot et al. (1996, 2001), this can happen due to the formation of soluble Cd-chloro complexes. It means that Cd will probably be a significant constituent of the drainage water as long as Cl<sup>-</sup> is present in sufficient concentrations to mobilize Cd.

The precipitation of Fe/Al-hydr(oxides) over the whole *L/S* range indicate that they were stable over time (Table 4). These minerals can be present in large amounts in weathered ashes, and they can adsorb trace elements such as As, Cd, Pb, Zn, Ni, Cu and Mo (Kumpiene et al., 2012, 2013; Meima and Comans, 1999, 2002; Zevenbergen et al., 1994). Thus, very low mobility of trace elements (Fig. 7) might be attributed to their adsorption by Fe/Al-hydr(oxides). However, higher release of trace elements may be expected if environmental conditions change towards acidic pH that favours a dissolution of Fe/Al hydr(oxides) and metal carbonates.

The higher release of organic matter in A4 compared to A1 was probably due to higher alkalinity of fine BA fraction compared to FA (Fig. 8). Easily leachable organic matter from the sludge and compost was likely to enhance the mobilization of Pb, Ni and Cu in the field from protection and vegetation layers since these elements showed positive correlation with TOC (correlation coefficients were 0.45–0.95). Since aerobic conditions are expected to retain in the upper parts of the cover, and if pH remains close to neutral in the pore water over time, Cu and Pb will probably be mobilized as long as soluble organic matter is available for leaching.

#### 4.3. Treatment needs for drainage water

The drainage water was contaminated with Cl<sup>-</sup>, N, TOC and many of the analysed trace elements (Table 2). Hence, the drainage water needs treatment before it can be discharged to the local recipient.

The portion of easily biologically degradable organic matter in the drainage water is assumed to be low since the sludge had been

digested before it was used in the construction. Also with regard to N and metals, the drainage water shows similar concentrations as methanogenic landfill leachate (Cerne et al., 2008; Lagerkvist, 2003). Therefore, the treatment of the drainage water could be done at a local leachate treatment plant, given that this facility contains all components that are necessary for treatment of the drainage water.

Biological treatment under oxidising and anoxic conditions may take care of the biologically degradable part of the organic matter and parts of the nitrogen while the metals could be reduced by a combined chemical and physical treatment step. Since the drainage water may contain high amount of  $\text{Cl}^-$  at low  $L/S$  ratios and total removal of  $\text{Cl}^-$  from drainage water might be very costly, the sea could be suitable recipient for the drainage water if the distance for pumping the water is reasonable.

Necessary treatment periods for the drainage water can be estimated taking into account the development of the drainage water quality in the laboratory tests, the initial  $L/S$  ratio and its increase of about  $0.171 \text{ (kg yr)}^{-1}$  as well as the local discharge criteria (Table 2). The estimated periods are quite different for different pollutants. For example,  $\text{Cl}^-$  will be depleted from the drainage water after about one-two decades after the cover was built (Fig. 6a). According to results from S1 (oxidising conditions), N will be depleted in the drainage water after a decade while a reduced environment (S2) can lead to periods of more than four decades before the discharge limits for N will be met (Table 2 and Fig. 6b). The development of concentrations of TOC shows that the drainage water will need treatment about three to four decades (Table 2 and Fig. 8). A low release of trace elements in S2 after  $L/S$  of  $4.21 \text{ (kg yr)}^{-1}$  indicates a rather short treatment period i.e. two decades (Table 2 and Fig. 7). Trace elements Cd, Cu, Ni and Zn were not immobilized in S1 after  $L/S$  of  $5.51 \text{ (kg yr)}^{-1}$  which indicates that the drainage water might be contaminated with these elements for at least three decades.

Pre-treatment of ashes, sludge and compost could be an option to minimize the treatment needs and periods for the drainage water. Chemical extraction and stabilization are possible pre-treatment methods for SCM to reduce the concentration of contaminants in the drainage water (Todorovic and Ecke, 2006). Information about costs for different pre-treatments of SCM could not be found in the literature, but a reasonable assumption is that pre-treatment costs will be a relevant part of any cover project and that they will vary depending on the local conditions such as material and treatment equipment availability, transport distances, energy cost etc. Thus, a comprehensive feasibility study about benefits of using SCM in a landfill cover, costs for pre-treatment of these materials and treatment needs for drainage water should be done before such materials can be used in landfill cover.

## 5. Conclusions

Using the SCM for the construction of a landfill cover affects the quality of the infiltrating water. The drainage water from the tested landfill cover built with ashes and sewage sludge was contaminated with  $\text{Cl}^-$ , N, TOC and several trace elements. The type and extent of the contamination depends directly on materials used, their composition, amounts (layer thickness), physical properties such as porosity, as well as leaching conditions such as pH, redox conditions, etc. The protection layer stands for the dominant impact on the drainage water quality while the vegetation and the drainage layer have less influence. The diffusion of trace elements from the liner surface is assumed to be low, but it may affect quality of drainage water after dry periods. Hence, the selection of the materials for the protection layer is crucial. The use of a sludge/FA mixture instead of a sludge/BA mixture in the protection layer may be a better option since it resulted in less contaminated drainage water.

A clear trend of all contaminants to decrease over time could be seen in the laboratory tests. Aging reactions in ashes such as carbonation and adsorption onto Fe/Al-hydr(oxides) probably decreased the release of trace elements from the ash-sludge mixture. Changes in the leaching conditions from a reducing towards an oxidizing atmosphere can result in longer release periods for trace elements from ashes (about one decade) but in a shorter release of N and organic matter from sludge (two–three decades). The solubility of salts and other trace elements from ashes is expected to be reduced over time also due to a decrease of temperature in the landfill cover following the stabilization of the landfilled waste.

Based on the concentration development of elements in the laboratory tests, it is estimated that the drainage water will need treatment for at least three to four decades after the completion of the landfill cover. Biological treatment combined with chemical/physical treatment of the drainage water is needed to remove organic matter, nitrogen and metals.

Overall, bearing in mind the reduced need of virgin construction materials, and environmental impacts associated with their production, the use of SCM can often yield an environmental benefit, and should be considered if available.

## Acknowledgements

This investigation was financially supported by Telge Återvinning AB and the Swedish Research Council for Environment, Agricultural Sciences and Spatial Planning (FORMAS). The authors thank Ulla-Britt Uvemo at the Department of Civil, Mining and Environmental Engineering, Luleå University of Technology, for conducting analyses and her patient assistance with the experiments. The authors are grateful to Dr Sofia Lidellöw, Luleå University of Technology and Dr Rolf Sjöblom, Tekedo AB, for their valuable comments.

## References

- Ali, H., Khan, E., Sajad, M.A., 2013. Phytoremediation of heavy metals – concepts and applications. *Chemosphere* 91, 869–881.
- Avfall Sverige, 2011. Förbättring av bottenaskors kvalitet, in Swedish, (Improvement of bottom ash quality). Report U2011:17. <[http://www.avfallsverige.se/fileadmin/uploads/Rapporter/Utveckling/Rapporter\\_2011/U2011-17.pdf](http://www.avfallsverige.se/fileadmin/uploads/Rapporter/Utveckling/Rapporter_2011/U2011-17.pdf)> (last access 2014-03-21).
- Bjurström, H., Lind, B.B., Lagerkvist, A., 2014. Unburned carbon in combustion residues from solid biofuels. *Fuel* 117 (PART A), 890–899.
- Brännvall, E., 2013. Suitability of Fly Ash for Construction and Land Applications. Doctoral Thesis, Luleå University of Technology. <[http://pure.ltu.se/portal/Lsv/publications/suitability-of-fly-ash-for-construction-and-land-applications\(27cd528f-cadc-42cf-98d8-8f7481bcd57\).html](http://pure.ltu.se/portal/Lsv/publications/suitability-of-fly-ash-for-construction-and-land-applications(27cd528f-cadc-42cf-98d8-8f7481bcd57).html)> (last access 2014-03-21).
- Brinkmann, U., Andreas, L., Conrad, H., Heyer, K.-U., Kabbe, G., 1999. Die Standard arbeits-anweisungen im BMBF-Verbundforschungs vorhaben "Deponiekörper". Müll und Abfall 31.
- CEN, 1997. Water analysis – Guidelines for the Determination of Total Organic Carbon (TOC) and Dissolved Organic Carbon (DOC), EN 1484. European Committee for Standardization (CEN), Brussels, Belgium.
- Cerne, O., Allard, A., Ek, M., Junestedt, C., Svenson, A., 2008. Utvärdering av behandlingsmetoder för lakvatten från deponier. in Swedish, (Evaluation of treatment methods for landfill leachate). IVL Svenska Miljöinstitutet, IVL Rapport B1748, Stockholm, Sweden.
- Chandler, A.J., Eighmy, T.T., Hartlen, J., Hjelmar, O., Kosson, D.S., Sawell, S.E., van der Sloot, H.A., Vehlow, J., 1997. Municipal Solid Waste Incinerator Residues. *Stud. Environ. Sci.* 67, Elsevier Science B.V., Amsterdam.
- Dijkstra, J.J., van der Sloot, H.A., Comans, N.J.R., 2006. The leaching of major and trace elements from MSWI bottom ash as a function of pH and time. *Appl. Geochem.* 21, 335–351.
- Gustafsson, J.P., Berggren Kleja, D., 2005. Modelling salt-dependant proton binding by organic soils with the NICA-Donnan and Stockholm Humic Models. *Environ. Sci. Technol.* 39, 5372–5377.
- Johnson, C.A., Brandenberger, S., Bassini, P., 1995. Acid neutralizing capacity of municipal waste incinerator bottom ash. *Environ. Sci. Technol.* 29, 142–147.
- Johnson, C.A., Kaeppli, M., Brandenberger, S., Ulrich, A., Baumann, W., 1999. Hydrological and geochemical factors affecting leachate composition in municipal solid waste incineration bottom ash. Part II. The geochemistry of leachate from Landfill Lostorf, Switzerland. *J. Contam. Hydrol.* 40, 239–259.

- Karlfeldt, K., Steenari, B.-M., 2007. Assessment of metal mobility in MSW incineration ashes using water as the reagent. *Fuel* 86, 1983–1993.
- Kidd, P., Barcelo, J., Bernal, M.P., Navari-Izzo, F., Poschenrieder, C., Shilev, S., Clemente, R., Monterroso, C., 2009. Trace element behavior at the root-soil interface: implications in phytoremediation. *Environ. Exp. Botany* 67, 243–259.
- Kosson, D.S., van der Sloot, H.A., Eighmy, T.T., 1996. An approach for estimation of contaminant release during utilization and disposal of municipal waste combustion residues. *J. Hazard. Mater.* 47, 43–75.
- Kumpiene, J., Lagerkvist, A., 2009. Referensdata för miljöbedömning av alternativa material i sluttäckning av avfallsupplag. – in Swedish (Reference values for environmental assessment of alternative materials in landfill covers), Report D2009:04. <<http://www.avfallsverige.se/fileadmin/uploads/Rapporter/Deponering/D2009-04.pdf>> (last access 2014-03-21).
- Kumpiene, J., Ragnvaldsson, D., Lövgren, L., Tesfalidet, S., Gustavsson, B., Lättström, A., Leffler, P., Maurice, C., 2009. Impact of water saturation level on arsenic and metal mobility in the Fe-amended soil. *Chemosphere* 74, 206–215.
- Kumpiene, J., Fitts, J.P., Mench, M., 2012. Arsenic fractionation in mine spoils 10 years after aided phytostabilization. *Environ. Pollut.* 166, 82–88.
- Kumpiene, J., Desogus, P., Schulenburg, S., Arenella, M., Renella, G., Brännvall, E., Lagerkvist, A., Andreas, L., Sjöblom, R., 2013. Utilisation of chemically stabilized arsenic-contaminated soil in a landfill cover. *Environ. Sci. Pollut. Res.* 12, 8649–8662.
- Kylefors, K., Andreas, L., Lagerkvist, A., 2003. A comparison of small-scale, pilot scale and large-scale tests for prediction behaviour of landfilled wastes. *Waste Manag.* 23, 45–59.
- Lagerkvist, A., 2003. Landfill technology. Technical Report 2003:15. Luleå University of Technology, ISSN: 1402–1536, Luleå, Sweden.
- Maurice C., Lagerkvist A., 1998. Utveckling av botten tätning för avfallsupplag baserad på vedaska – In Swedish (Development of bottom liner for landfills based on bio ash), Stiftelsen Reforsk, p. 146.
- Meima, J.A., Comans, R.N.J., 1998. Application of surface complexation/precipitation modelling to contaminant leaching from weathered municipal solid waste incinerator bottom ash. *Environ. Sci. Technol.* 32, 688–693.
- Meima, J.A., Comans, R.N.J., 1999. The leaching of trace elements from municipal solid waste incinerator bottom ash at different stages of weathering. *Appl. Geochem.* 14, 159–171.
- Meima, J.A., Comans, R.N.J., 2002. Carbonation processes in municipal solid waste incinerator bottom ash and their effect on the leaching of copper and molybdenum. *Appl. Geochem.* 17, 1503–1513.
- Petrucci, R.H., 1985. *General Chemistry. Principles and Modern Applications.* Macmillan Publishing Company, New York.
- Raab, B., Vedin, H., 1995. *Climate, lakes and rivers.* National Atlas of Sweden, vol. 14. Stockholm, Sweden.
- Sabbas, T., Poletini, A., Pomi, R., Astrup, T., Hjelmar, O., Mostbauer, P., Cappai, G., Magel, G., Salhofer, S., Speiser, C., 2003. Management of municipal solid waste incineration residues. *Waste Manag.* 23, 61–88.
- SFS., 2001. Förordning (2001:512) om deponering av avfall (Ordinance on the landfilling of waste), SFS (Svensk författningssamling, Swedish statute book) no: 2001:512 (in Swedish). Miljödepartementet (Ministry of the Environment), Stockholm, Sweden.
- SIS., 1976. Determination of nitrogen compounds in water. Oxidation with peroxodisulphate, SS 028131. Swedish Standards Institute (SIS), Stockholm, Sweden.
- Tao, S., Liu, W.X., Chen, Y.J., Xu, F.L., Dawson, R.W., Li, B.G., Cao, J., Wang, X.J., Hu, J.Y., Fang, J.Y., 2004. Evaluation of factors influencing root-induced changes of copper fractionation in rhizosphere of a calcareous soil. *Environ. Pollut.* 129, 5–12.
- Todorovic, J., Ecke, H., 2006. Treatment of MSWI residues for utilization as secondary construction minerals: a review of methods. *Miner. Energy* 20, 45–59.
- Travar, I., Lidelöv, S., Andreas, L., Tham, G., Lagerkvist, A., 2009. Assessing the environmental impact of ashes used in landfill cover construction. *Waste Manag.* 29, 1336–1346.
- van der Sloot, H.A., 1996. Developments in evaluating environmental impact from utilization of bulk inert wastes using laboratory leaching tests and field verification. *Waste Manag.* 16, 65–81.
- van der Sloot, H.A., Comans, R.N.J., Hjelmar, O., 1996. Similarities in leaching behavior of trace contaminants from waste, stabilized waste, construction materials and soils. *Sci. Total Environ.* 178, 111–126.
- van der Sloot, H.A., Kosson, D.S., Hjelmar, O., 2001. Characteristics, treatment and utilization of residues from municipal waste incineration. *Waste Manag.* 21, 753–765.
- Yoon, J., Cao, X., Zhou, Q., Ma, L.Q., 2006. Accumulation of Pb, Cu and Zn in native plants growing on a contaminated Florida site. *Sci. Total Environ.* 368, 456–464.
- Zevenbergen, C., Bradley, J.P., van der Wood, T., Brown, R.S., van Reeuwijk, L.P., Schuiling, R.D., 1994. Natural weathering of MSWI bottom ash in a disposal environment. *Microbeam Anal.* 3, 125–135.
- Zevenbergen, C., van Reeuwijk, L.P., Bradley, J.P., Bloemen, P., Comans, R.N.J., 1996. Mechanism and conditions of clay formation during natural weathering of MSWI bottom ash. *Clays Clay Miner.* 44, 546–552.