DEVELOPMENT OF DRAINAGE WATER QUALITY FROM LANDFILL COVERS BUILT WITH ASHES AND SEWAGE SLUDGE

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SUMMARY: An alternative to virgin and artificial materials in landfill cover can be various types of waste materials like ashes and sewage sludge. From the environmental point of view, the most interesting question to study is the quality of the drainage water generated above the liner in landfill cover built with waste materials. Thus, the main aim of this paper is the evaluation of drainage water quality and time required for its treatment. Results from a full scale test, physical models and a column test were used in the evaluation of the drainage water quality development. The main contaminants identified in drainage water were N-tot, As, Cu, Cd, Pb, Zn and Ni. The laboratory tests showed higher concentrations of N-tot, NH4-N, TOC, As and Pb, but lower values of Cl, Zn, Ba and Ni in comparison with drainage water from the field. It was probably a result of different factors that may have influence on leaching but interactions of the protection layer with the vegetation layer and the liner surface are expected to influence the results from the field. According to results from the laboratory experiments, drainage water will probably need treatment for at least two-three decades after covering.

1. INTRODUCTION

Many old landfills in both EU and Sweden will be closed in the nearest future. According to Swedish legislation, terminated municipal and hazardous waste landfills must be capped. It is estimated that about 50 000 t ha^{-1} of natural materials is needed in landfill cover construction. This will result in a costs about 2 million Euros ha^{-1} and in a strain to the environment through the exploitation of natural materials. An alternative can be the utilization of various waste materials i.e. secondary construction materials (SCM) like ashes, sludge, sand from fluid bed incinerators and compost. These materials must meet the same functional requirements as the materials they replace and sufficient quantities must be available on the market. However, use of wastes in constructions raises serious environmental concern due to potential leaching of for example trace elements, nutrients and organic compounds.

A landfill cover is a multilayer construction that serves to reduce infiltration of water to deposited waste and to minimize emission of landfill gas into the atmosphere. Two types of water may be generated in a landfill cover. The leachate corresponds to water that percolates through the liner, while the drainage seeps through the landfill cover layers above the liner. Given that the design is reasonable, most of the infiltrated water will leave the landfill cover as drainage water. Thus, from an environmental point of view, placement of SCM above the liner is more critical than within or below the liner.

In order to test different SCM in landfill cover, a six hectares large test area was established at the Tveta landfill, about 40 km southwest of Stockholm, Sweden. At Sardinia 2005 and 2007, a description of the design and construction of the test area, description of measurement equipment and preliminary results of leachate and drainage water quality were presented (Travar *et al.*, 2005, Travar *et al.*, 2007). The environmental impact of drainage and leachate quality from landfill cover was studied in detail (Travar *et al.*, 2009).

The overall aim of this study is to assess the need of and estimate the time required for treatment of drainage water from the landfill cover. Physical models (hereafter simulators) and column tests were used to simulate drainage water quality development. Together with field data from more than four years these results were evaluated to assess if development of drainage quality under field conditions can be predicted from laboratory tests.

2. MATERIAL AND METHODS

2.1 Material characterization

Figure 1 shows the material mixtures used in the full scale experiments at the Tveta landfill. Before the materials were used in the construction a one-step batch leaching test at a liquid to solid ratio (L/S) of $10 \, l \, kg^{-1}$ was used to estimate the water soluble fraction of compounds in the materials. The results from the test were used to assess acceptance of material before it was applied in the construction and to evaluate long-term leaching. In order to estimate the development of drainage water quality from the landfill cover, material of the protection layer from area 4 (A4) taken during construction works was used in the simulator test (described below). Moreover, excavated samples taken from the middle of the protection layer from areas 1 (A1) and 4 (A4) after four and two years since landfill cover was constructed were used in the column test (Table 1). In order to study the transport of different elements/components through the landfill cover, samples of the protection layer, drainage layer and liner material were excavated from A1 and A4 (results are not shown in this paper). Table 1 summarizes the tested materials and leaching tests used for the assessment of drainage water quality.

A 7   A 6   A 4   A 1

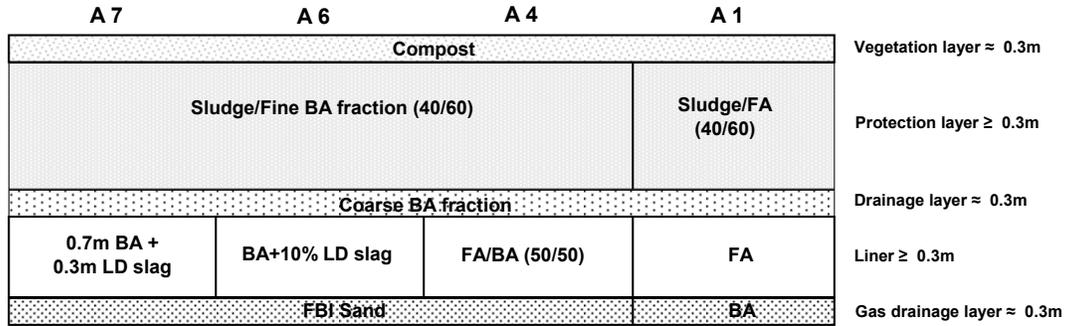| Compost | | | | Vegetation layer ≈ 0.3m |
Sludge/Fine BA fraction (40/60)			Sludge/FA (40/60)	Protection layer ≥ 0.3m
Coarse BA fraction				Drainage layer ≈ 0.3m
0.7m BA + 0.3m LD slag	BA+10% LD slag	FA/BA (50/50)	FA	Liner ≥ 0.3m
FBI Sand			BA	Gas drainage layer ≈ 0.3m

Figure 1 Design of the sub-areas A1, A4, A6 and A7 of the landfill cover test area at the Tveta landfill. BA-bottom ash, FA-fly ash, FBI-fluid bed incineration

Table 1. Overview of excavated materials and applied tests

Material	Area	Description	Sampling occasion	Batch test L/S=10	Column test	Simulators
Sludge+BA	4	Protection layer	During construction works			X
Sludge+BA	4	Protection layer	Excavated after 2 years, taken every 20 cm of layer depth	X		
Sludge+BA	4	Protection layer	Excavated after 2 years, taken from the middle of the layer		X	
Sludge+FA	1	Protection layer	Excavated after 4 years, taken every 20 cm of layer depth	X		
Sludge+FA	1	Protection layer	Excavated after 4 years, taken from the middle of the layer		X	
BA	4	Drainage layer	Excavated after 2 years, taken from the middle of the layer	X		
BA	1	Drainage layer	Excavated after 4 years, taken from the middle of the layer	X		
FA	1	Liner	Excavated after 4 years, taken from the surface	X		
FA/BA (50/50)	4	Liner	Excavated after 2 years, taken from the surface	X		

2.2 Batch leaching test

A one-step leaching test at L/S=10 was used to evaluate the release of water soluble fraction of elements from the samples. The test was run for 24 hours using an end-over-end tumbler.

2.3 Column leaching test

The column leaching test was performed in triplicates according to standard SIS-CEN /TS 14405 (up-flow percolation test). Material from the middle of the protection layer of areas 1 and

4 was studied in this test.

2.4 Simulators

Two cylindrical stainless steel containers with a total volume of about 100 litres (ø 39 cm, height 84 cm) were used to simulate the development of drainage water that percolates through the protection layer (Figure 2). These simulators are designed to carry out events under real conditions on a laboratory scale. An acceleration of chemical, biological and physical processes can be caused by modified water balances and optimal conditions for microorganisms, like constant temperatures and homogeneous moisture distribution. The volume of material in each simulator was about 70 litres. The simulators were connected in a series. The first simulator was open to the atmosphere simulating the upper part of the protection layer. The second simulator was hermetically closed simulating the lower part of the protection layer. Every fortnight 5 litres of tap water were added to the first simulator. After 48 h it was pumped to the second simulator. The retention time for the water in the second simulator was about two weeks after which it was discharged and left to rest for a week. The simulators were kept indoors at about 20°C. Drainage water samples were taken from both simulators continually after about each L/S 0.15 and kept frozen at -20 °C prior analysis. In this test, the L/S ratio was calculated using the amount of added and infiltrated water. The L/S 2.65 was reached in this experiment after about two years.
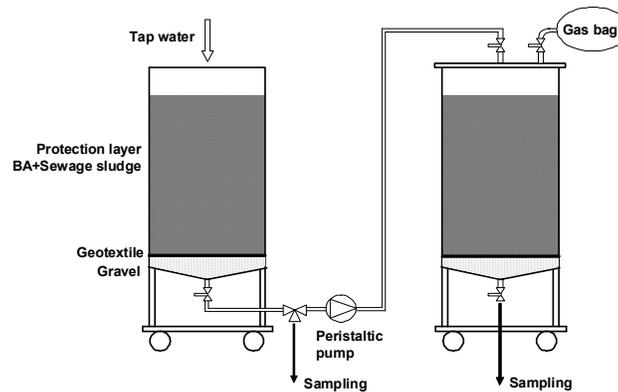

Figure 2. Design of the simulators filled with protection layer material

2.5 Full scale test

The design of the test areas and description of the measurement equipment was presented at Sardinia 2005 and 2007 (Travar *et al*. (2005), Travar *et al*. (2007), Travar *et al*, 2009). The drainage water from areas 6 and 7 was collected at one sampling point, i.e. drainage 6+7 is a mixture of the drainage water that comes from areas 6 and 7.

The L/S ratio in the field test was calculated using precipitation data and assumption that 30% of total precipitation accumulates as drainage water. It is calculated that the L/S 0.65 was reached in A1 5.5 years after covering. The L/S 0.51 is calculated for A4 and L/S 0.46 is calculated for A6+7 that were reached after 4 and 3.5 years respectively since construction was finished.
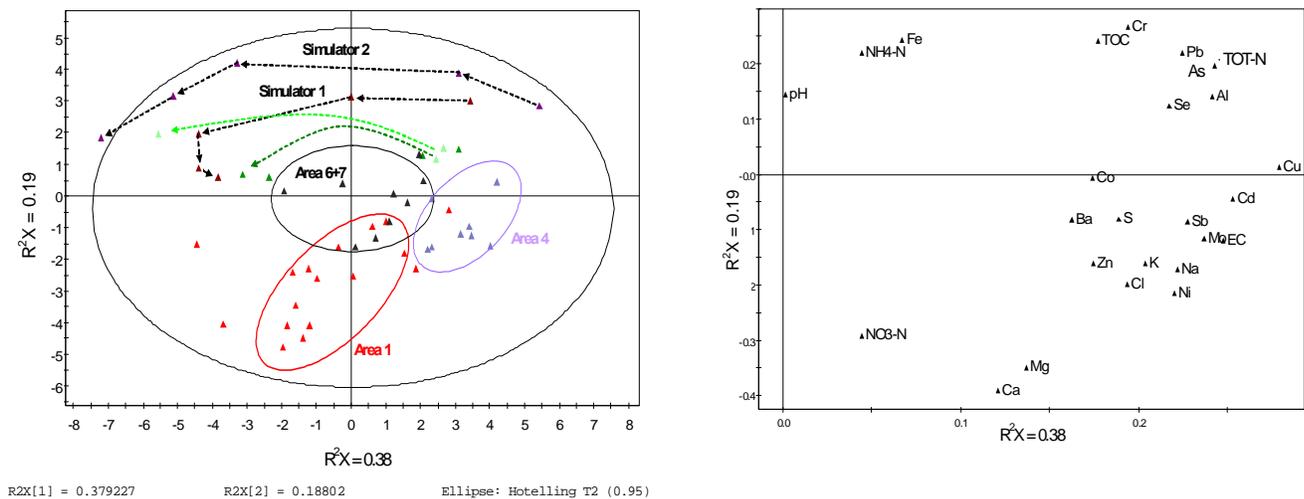
2.6 Analyses

Aqueous samples were analysed for elemental concentrations using ICP-SFMS (EPA method 200.8; modified) without prior digestion. Cl$^-$ was analysed spectrophotometrically (AACE Quaatro, Bran+Luebbe, Germany). Total nitrogen (N$_{tot}$) and ammonia nitrogen (NH$_4$-N) were analysed using AACE Quaatro autoanalyser (Bran+Luebbe, Germany). N-tot was analysed after digestion according to the Swedish standard method SS 028131 (SIS, 1976). Total organic carbon (TOC) was determined according to EN 1484 (CEN, 1997). Analysis of most of the elements was accurate within 20% analytical error.

3. RESAULTS AND DISCUSSION

3. 1 Drainage water quality in the field

An overview of the drainage water quality from the field, simulators and column leaching test is presented in principal component analysis (PCA) score and loading plots (Figure 3). These plots are complementary and a direction in one plot corresponds to the same direction in the other plot. The variables positioned further away from the plot origin have a stronger impact on the model and the placement also shows the correlation between variables. For example, the elements Na, K and Cl are strongly correlated with regard to both principal components, if Cl decreases; the same is valid for Na and K. The ellipse defines the 95% confidence interval in the score plot. The degree of explanation for the first two principal components is 57% of the data variation.

Drainage water samples from the field are clearly grouped according to the material used in the protection layer but also in the liner (Figure 3). The dominant contaminants in drainage water from A4 were Cu, Cd, As, Pb and N-tot, while drainage from A1 contained the highest amounts of Zn, Ni and Cl in comparison with drainage from other areas (Figure 3). The quality of drainage from A6+7 was between that of A4 and A1. Since the same material mixture is used in the protection layer of A4 and A6+7, the difference between these areas is probably due to the influence of the liner's surface when drainage water flows over. Moreover, it could be also a result of the differences in the composition of the sludge/BA mixture since these areas were not covered at the same time.
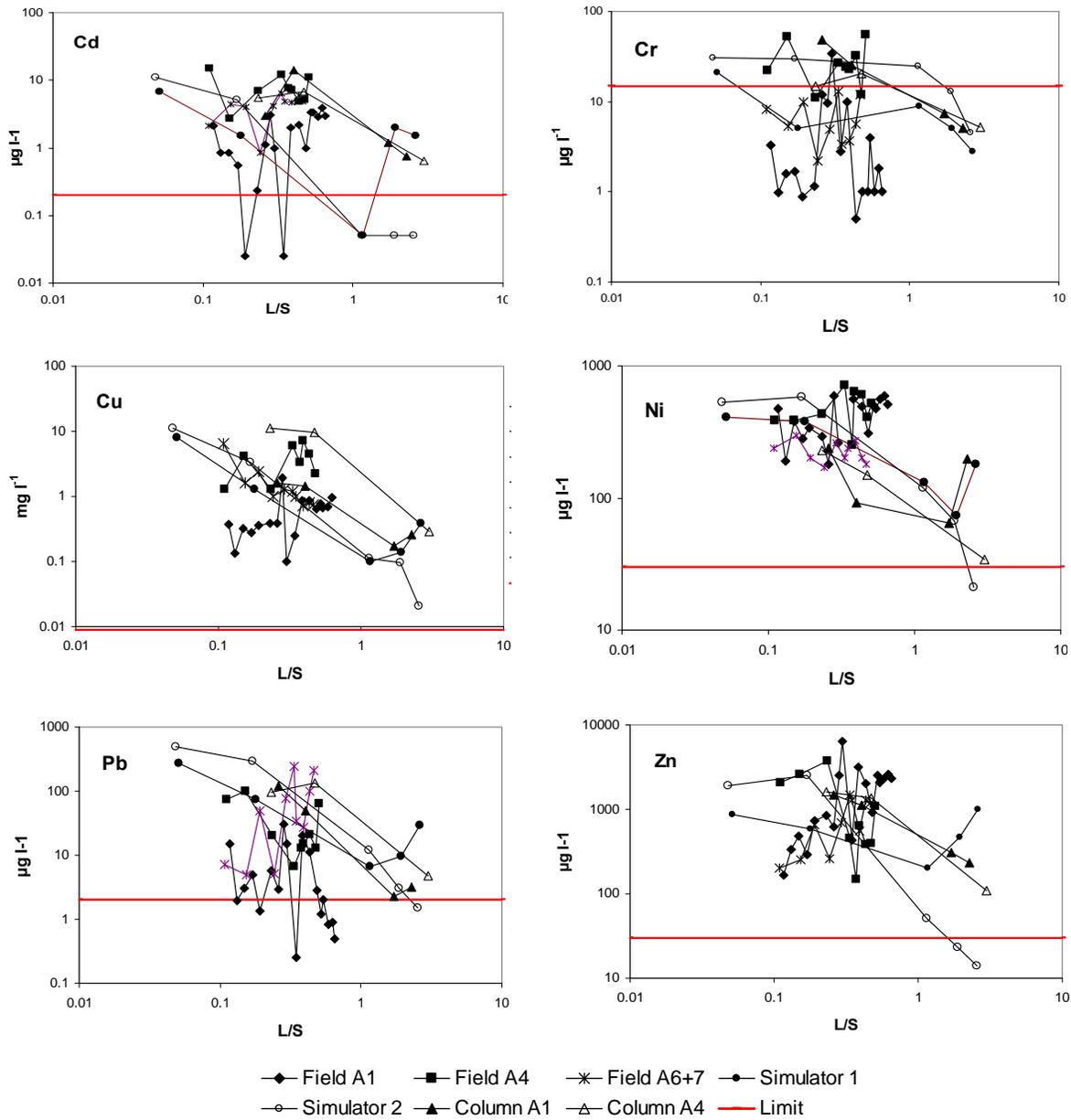
Figure 3. An overview of the drainage water quality from the field, simulators and column experiment presented in scatter (left) and loading (left) plot. The arrows represent moving of responses from the simulator test and the column test with increasing L/S.

Figures 4 and 5 illustrate changes in concentrations of components and elements over L/S in the drainage water from the field, simulators and column test. For the presented elements the landfill operator has to meet certain demands for discharge to the recipient. The straight lines in the figures represent the limit concentration for each element (no limit is set for chloride). The concentrations of the drainage water from the field exceed the limit for all elements except for NH4-N in areas 1 and 6+7, Cr in all areas and As, Pb and Cd in area 1. The concentrations of Cu and Zn were two orders of magnitude higher than the limit values while Ni, Pb Cd, and N-tot showed one order of magnitude higher values in comparison with limit values. Thus, drainage water from all areas needs treatment before it can be discharged to the recipient.

Figure 4 Changes in concentrations of N-tot and NH4-N with L/S in drainage water from the field, simulators and column tests. The straight line represents the limit value for direct discharge to the recipient.

Figure 5 Changes in concentrations of Cl⁻, As, Cd, Cr, Cu, Ni, Pb and Zn with L/S in drainage water from the field, simulators and column tests. The straight line represents the limit value for direct discharge to the recipient.

3.2 Field versus laboratory experiments

Comparing the results from the lab tests with the field data, the initial values from the column test are positioned close to the drainage water quality from area 4 and 6+7 but further away from area 1 while the results from the simulator test are positioned further away from all field data (Figure 3). The higher concentrations of almost all elements/components in the simulator test can be explained by the fact that the first sample from the simulator test was taken at a lower L/S

ratio than from both column and field test (Figure 4 and 5). However, the laboratory tests showed higher concentrations of N-tot, NH4-N, TOC, As and Pb, but lower values of Cl, Zn, Ba and Ni in comparison with drainage water from the field (Figure 3, 4 and 5). One reason for these differences could be the mobilization of constituents from the liner surface or influences from the vegetation layer. Another explanation for the differences between drainage quality from the laboratory and field test are differences in the liquid to solid (L/S) ratio, biological degradation, duration of the experiment, size of the particles, temperature and sample preparation/storage (Kylefors *et al.*, 2003).

The higher Cl$^-$ content in the drainage water from the field test in comparison with laboratory experiments could be a result of an erroneous assumption about the water infiltration in the field. Another explanation could be damage of the construction which resulted in a lower flow of drainage water to the sampling points.

As a result of biological processes in the sludge, pH and redox potential are affected which in turn influence the leaching behaviour. For example, the difference in drainage pH between the field test and simulators can be a reason for different releases of Pb, Zn, Cd and Cu (van der Sloot, 2001). The pH values in the field test varied between 7 and 8.9 while the laboratory tests showed pH between 6.9 and 8.1 (Figure 6). The difference between pH in the field test and laboratory tests was not found to be significant ($\alpha = 0.05$). Thus, the different release of Pb and Zn in the field test and in the laboratory tests can not be explained at this stage. Redox conditions are likely to influence the release of redox sensitive elements such as Cu, As, Cr and Fe from the protection layer material. Similar concentrations of Fe were observed in both simulators (results not shown) which is probably a result of the similar redox conditions in both simulators. Concentrations of Fe in the field were one to two orders of magnitude lower than in simulators which may imply that redox was more reduced in the simulator test in comparison with the field test.
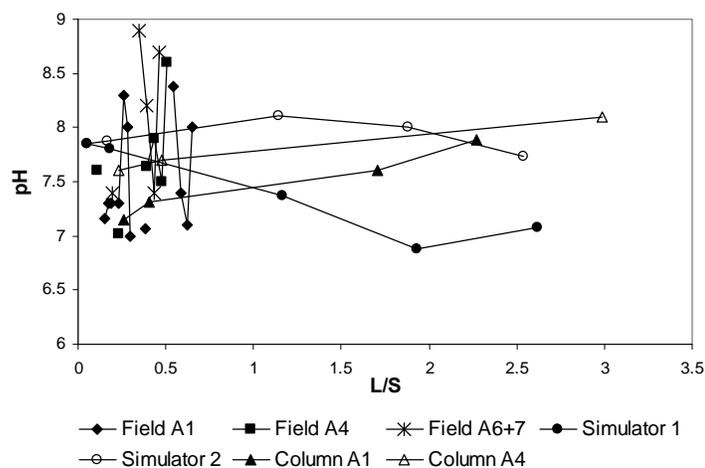

Figure 6. Development of pH in the field (Field A1, A4 and A6+7), simulators and column experiment (Column A1 and A4)

The duration of the experiment can affect the chemical equilibrium between solid and liquid phase and enable the establishment and continuation of biological reactions. An optimal retention time for reaching chemical equilibrium in experiments with acidogenic biological degradation is 12-24 h (Kylefors *et al.*, 2003 after Pavan *et al.*, 2000). Thus, two weeks retention time in the

simulators should be more than enough in order to reach equilibrium conditions similar to the field.

Temperature may affect chemical equilibrium and biological activity in the protection layer and thus release of elements from the solid phase. Measured temperatures in the middle of the protection layer in all areas were between 10 °C in the winter and 25 °C in the summer period (results not presented). It is assumed temperature has minor influence on different leaching conditions between the laboratory and the filed test since temperature in laboratory experiments was about 20°C.

All in all, mobilization/immobilization of elements in the field compared to the laboratory experiments is probably a result of different factors that may influence leaching. Moreover, the liner surface and vegetation layer may contribute to the drainage water quality which was not simulated in the laboratory tests.

3.3 Development of drainage water quality

The L/S can be used as a tool for comparing results from simulators and column tests with the full scale test. It is a more objective tool than time because it takes into account the amount of solid material and water that percolates through the construction. However, using the L/S ratio to predict for example discharge concentrations for drainage has limitations because it does not consider kinetics or changes in environmental conditions. The prediction of required L/S to reach a certain level of concentration is also associated with great uncertainties in simulators. For example, Kylefors *et al*. (2003) stated that the uncertainty in predictions is associated with a factor of at least 4. However, the simulator experiments can give useful predictions if degradation is proceeding in the same phase (Kylefors *et* al., 2003).

Responses from simulators and column test move to the left in score plot (Figure 3) which is a result of the lower release of elements with increasing L/S, i.e. all presented elements in lab tests showed decreasing trend with increasing L/S ratio (Figure 4 and 5). However, the concentrations of the most elements in drainage water from the field do not show tendency to decrease after couple of years since landfill cover was built. For example, the concentrations of N-tot, NH4-N, Cd and Cu in drainage water of area 1 showed decreasing trend in the beginning but the concentrations increased again when L/S reached above 0.5. Zn showed increasing concentrations in areas 1 and 6+7 which were opposite to the results of the laboratory experiments. Elements like As, Cr, Ni and Pb and ion Cl⁻ showed varying concentrations in all areas with increasing L/S. Thus, the achieved L/S of 0.65 in the field test was not enough to observe any clear decrease in concentrations of elements in the field.

Results from the simulators and column test showed that N-tot, Cd, Cu and Zn have not reached discharge limits with L/S of 2.5 i.e. the concentrations of these elements in drainage water will probably still be higher than limit values after a couple of decades after construction of the landfill cover. The upper part of the protection layer (simulator 1) showed concentrations of N-tot of about 40 mg l^{-1} but the lower part (simulator 2) points at concentrations of about 370 mg l^{-1} after L/S 2.5. The concentration of Cu was quite similar in both simulators except for the last measurement at L/S of 2.5. At this point, concentrations of Cu in simulator 2 and 1 were about two and 40 times higher than the limit value, respectively. Cu forms complexes with dissolved organic matter and it is transported to the liquid phase as water soluble organic complex (van der Sloot et al, 2001; Chandler et al., 1997). As it was mentioned before, simulator test can not simulate kinetics of degradation reactions as they occur on a full scale under different environmental conditions over time. However, it is assumed that the most of the ash/sludge-mixtures will be probably be degraded within a few decades. Thus, the release of Cu from the layers above the liner might decline considerably over time. After L/S 1, concentration of Cd was

under detection limit in the simulator 2 but it was still available for leaching in simulator 1 and the column test. Cd showed positive correlation with Cl meaning that lower release of Cl was followed by lower concentrations of Cd. Thus, further decrease of Cl with increasing L/S over 2.5 will be probably result in Cd concentrations in simulator 1 and column test below limit value. Release of Zn differed between simulators while the column test showed similar release of Zn between areas 1 and 4. The pH values in the two simulators were about the same (Figure 6) and, hence, the different release of Zn from simulators can not be attributed to pH. According to the results from simulator 2, Zn in drainage water will reach the discharge limit after about L/S 1.5 while the column test showed one order of magnitude higher concentrations than limit after about L/S 2.5. Thus, it is assumed that drainage water will probably need treatment with regard to nitrogen, Cu, Cd and Zn for several decades after the landfill cover was built.

4. CONCLUSIONS

Drainage water samples in the field test were mainly contaminated with Cu, Cd, As, Pb, Zn, Ni and N-tot. Thus, drainage water needs treatment before it can be discharged to the recipient. The laboratory tests showed higher concentrations of N-tot, NH4-N, TOC, As and Pb, but lower values of Cl, Zn, Ba and Ni in comparison with drainage water from the field. It was probably a result of different factors that may have influence on leaching but interactions of the protection layer with the vegetation layer and the liner surface are expected to influence the results from the field.

An achieved L/S of 0.65 in the field was not enough to observe clear trend for decreasing concentration of elements. However, results from the laboratory tests show clear trend of all elements to decrease. In the simulator test, concentrations of N-tot, Cd, Cu and Zn did not reach the limit value for discharge to the recipient after L/S 2.65. Thus, it is assumed that drainage water will probably need treatment at least couple of decades after construction of landfill cover.

More results, especially for N-tot, Cd, Cu and Zn, are needed from the laboratory tests to make better assessment about drainage water quality development. Moreover, geochemical modelling can be used to identify mineral phases that control the release of elements and minerals that can precipitate with increasing L/S. Also, further analyses of the excavated samples from the landfill cover may contribute to a better understanding of the transport processes in and long-term emissions from the protection layer.

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